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RICE UNIVERSITY

Design and Development of an Apparatus to Study Atom-Surface Interactions Using Rydberg Atoms

by

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ABSTRACT

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An apparatus has been developed for studying the interaction of Rydberg atoms with surfaces, particularly the perturbation of Rydberg states by a nearby surface and the distance from the surface at which resonance ionization occurs. For this purpose, a source of metastable Xe(3P0,2) atoms was constructed and characterized. Using single photon laser excitation from the 6s' [1/2]o state, the nf[3/2], Rydberg series for n=16-20 has been observed with signals well above the background. Measurements of the energy levels of these Rydberg states has identified systematic errors in quoted energy levels from earlier work. The isotopic structure of the 20f Rydberg state was resolved, and the effect of Rydberg excitation in an electric field was investigated. Studies are underway to better understand the behavior of Xe(nf) Rydberg atoms in an electric field. This information will be used in subsequent experiments to study Rydberg-surface interactions.
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# Table of Contents

Abstract .................................................................................................................................................. ii  
Acknowledgements ............................................................................................................................... iii  
List of Figures .......................................................................................................................................... v  
**Chapter 1: Introduction** .................................................................................................................. 1  
**Chapter 2: Theory** ............................................................................................................................ 9  
**Chapter 3: Experiment** .................................................................................................................... 17  
  A. General Overview .......................................................................................................................... 17  
  B. Metastable Atom Source .............................................................................................................. 20  
  C. Characterization of Source Performance ..................................................................................... 22  
  D. Rydberg State Excitation .............................................................................................................. 27  
  E. Laser System ................................................................................................................................. 30  
  F. Main Interaction Region ................................................................................................................. 32  
**Chapter 4: Results and Future Direction** ....................................................................................... 39  
**References** ....................................................................................................................................... 50
List of Figures

Fig 1.1 Overview of present approach .............................................. 3
Fig 1.2 \( n=8 \) Stark states ............................................................... 6
Fig 1.3 Linear Stark effect ............................................................... 7
Fig 2.1 \( H(n=10), m=0 \) energy levels and widths vs. \( Z \) ................. 11
Fig 2.2 \( H(n=10), m=0 \) probability densities ..................................... 12
Fig 2.3 Ion production and minimum electric field vs. \( Z \) ................... 14
Fig 3.1 Schematic diagram of apparatus ............................................. 19
Fig 3.2 Schematic diagram of \( \text{Xe}(^3P_0) \) metastable atom source ........ 21
Fig 3.3 Stern-Gerlach profiles for \( \text{Ar} \) and \( \text{Xe} \) metastable beams ...... 26
Fig 3.4 Xenon energy level diagram ................................................... 28
Fig 3.5 Power tuning curves for Coumarin 47 and Coumarin 102 .......... 31
Fig 3.6 Schematic diagram of Main Interaction Region ....................... 33
Fig 3.7 Background electron count rates for MIR ............................... 36
Fig 3.8 Background ion count rates for MIR ..................................... 37
Tbl. 4.1 \( 6s' \rightarrow nf \) transition wavelengths, estimated and measured ....... 40
Fig 4.1 Quantum defect vs. \( n \), calculated and measured ...................... 41
Fig 4.2 Ion signal vs. laser frequency near \( n=20 \) transition,
\hspace{1cm} E=2V/cm and E=20V/cm ...................................................... 43
Fig 4.4 Ion signal vs. laser frequency near \( n=20 \) transition,
\hspace{1cm} E=50V/cm ..................................................................... 45
Fig 4.4 Count rate vs. laser power for \( n=20 \) ................................. 46
Chapter 1: Introduction

The interaction of individual or beams of particles with various surfaces presents an important problem in modern physics. While much has been learned about the interaction of photons, electrons, ions, and neutral metastable atoms with surfaces, a relatively neglected area of study is the behavior of highly excited (Rydberg) atoms at surfaces. Because of its large physical size and weak binding energy, a Rydberg atom is expected to be strongly perturbed by a nearby conducting surface. At a critical distance from the surface, the excited Rydberg electron should tunnel into the conductor; a process known as "resonance ionization." By determining the atom-surface separation at which resonance ionization occurs, one can obtain information on the electronic potential at relatively large (hundreds of angstroms) distances from the surface. Such measurements can provide new insights into electron tunneling processes at surfaces, which form the basis of many practical surface spectroscopies. This thesis discusses the design and development of an apparatus for studying the interaction of xenon Rydberg atoms with a metal surface.

A. General Properties of Rydberg atoms

In short, any atom or molecule in which the outermost electron is excited to a state of large principal quantum number \( n \) is said to be in a Rydberg state. Many fundamental properties of Rydberg atoms depend critically on \( n \). The size of the atomic electron cloud increases rapidly with \( n \) and is approximated by \( r_n = n^2a_0 \), where \( a_0 = 5.3 \times 10^{-9} \text{cm} \) is the Bohr radius of the ground state Hydrogen atom. One can see that even exciting Hydrogen to an \( n=10 \) state dramatically increases the physical size of the atom. Because the excited electron is far from the nucleus, it experiences a
nearly Coulombic potential. Therefore, to first order, a high \( n \) state of one element behaves much like the same high \( n \) state of a different element.

The binding energy of the excited electron is, to a good approximation, determined by \( E_{n,l} = R/(n-\delta_l)^2 \), where \( \delta_l \), called the quantum defect, depends on the orbital angular momentum quantum number \( l \) of the excited electron and the element in question. \( R \) is the Rydberg constant and is element specific. The binding energy of an excited electron decreases rapidly with increasing principal quantum number. The excited electron in a Rydberg atom is thus loosely bound to the core ion.

These physical characteristics suggest that Rydberg atoms will behave in novel ways near a conducting surface. Because of the atom’s large size, the Coulomb attraction between the excited electron and the core ion is small for a Rydberg atom. Near a surface, then, image charge effects are expected to strongly perturb the excited electron’s orbit. Indeed, at relatively large core ion-surface distances, the image charge interaction with the Rydberg electron can dominate the core ion Coulomb attraction, and the Rydberg atom will ionize.

The other general property of a Rydberg atom relevant to this experiment is its relatively long lifetime. For low \( l \) states, the lifetime of a Rydberg atom scales as \( n^3 \).\(^1\) The lifetime of a \( \text{Xe}(20f) \) Rydberg state is \( \sim 2 \mu s \).\(^2\)

**B. General Experimental Approach**

The goal of the present experiment is to determine the distance at which resonance ionization occurs when \( \text{Xe} \) Rydberg atoms are incident on a conducting surface. A schematic drawing of the present approach is shown in figure 1.1. A beam of metastable \( \text{Xe} \) atoms is directed toward a flat copper surface at near grazing incidence. Millimeters from the surface,
Figure 1.1  Schematic diagram of the present approach.  (a) The Rydberg atom approaches the surface at a shallow angle.  The electron is attracted to the surface by its image charge and tunnels into the conductor, leaving a core ion. (b) The ion is now attracted to the surface by its own image charge. At the surface, it will be neutralized by an electron from the conductor. (c) An electric field is applied to counteract the image charge attraction of the ion. This electric field directs the ion to a charged particle detector. The critical electric field necessary for just pulling the ion away from the surface is related to the distance from the surface at which resonance ionization occurred.
a fraction of the Xe metastables are photo-excited to a Rydberg state. As the Rydberg atom approaches the surface, the excited electron experiences an increasing attraction toward the surface from its positive image charge. At a critical distance, this image charge attraction overcomes the relatively weak Coulomb attraction between the electron and the core ion. The electron then tunnels into the conduction band of the surface (resonance ionization) and leaves behind the Xe⁺ core.

The resulting positive ion is now attracted to the surface by its own negative image charge. Close to the surface (a few angstroms), an electron will tunnel from the surface into the ion, leading to neutralization. If the ion neutralizes, the distance at which resonance ionization occurred cannot be determined. Therefore, an electric field is applied to pull the product ions away from the surface and toward a charged particle detector. This electric field must be strong enough to overcome the image charge attraction of the ion with the surface and redirect the small component of the Rydberg atom’s velocity perpendicular to the surface. The critical electric field necessary for just pulling the ion away from the surface can be related to the ion-surface distance at which resonance ionization occurred and the initial perpendicular velocity. By increasing the applied electric field until an increase in signal is seen on the charged particle detector, the distance for resonance ionization can be determined.

C. Rydberg Atoms in an Electric Field.

Because the experiment must be performed in the presence of an electric field, a detailed understanding of the behavior of Rydberg atoms in an external electric field is necessary for the success of the experiment. Much work, both theoretical and experimental, has been done to understand
the behavior of Rydberg atoms in an electric field.\textsuperscript{13} I will only briefly summarize this behavior here.

If a uniform electric field is applied in the $z$ direction, the hydrogenic potential becomes

$$V = -\frac{1}{r} + Ez,$$  \hfill (1.1)

using atomic units. The Schrödinger equation for an electron in this potential is no longer separable in spherical coordinates. For small values of $E$, one may treat the second part of the right hand side as a perturbation to the zero field states, $|n,l,m>$, in order to determine the Stark shift in energy caused by the electric field. Once this energy shift becomes comparable to the separation in energy between adjacent $l$ states this approximation is no longer valid, and $l$ is no longer a good quantum number. However, the Schrödinger equation is separable in parabolic coordinates for this potential, with the states now characterized by the quantum numbers $n$, $n_1$, $n_2$, and $m$. Using the parabolic basis to determine the perturbation caused by the electric field is straightforward. Figure 1.2 from Kleppner \textsuperscript{4} shows the $n=8$, $m=0$ parabolic states of hydrogen in an electric field.

The electric field not only affects the outermost electron's probability distribution, but the different Stark states now have different energies that are field dependent. Figure 1.3 shows the first order effect of an increasing electric field on the energies of the $n=10$ hydrogen Stark states. These energy shifts are associated with the classical dipole moment of the atom interacting with the electric field. The state with the largest dipole moment anti-parallel to the field ($n_1-n_2=-7$ in figure 1.2) is highest in energy, and that with the largest moment parallel ($n_1-n_2=7$ in figure 1.2) is lowest in energy. The energy separation between each Stark level increases with field. One can imagine this happening to every $n$ level for the atom, until the highest
Figure 1.2  Charge distributions for the eight $n = 8$ states of the hydrogen atom in an electric field. The effect of the electric field is quite pronounced. (Taken from Kleppner\textsuperscript{4}).
Figure 1.3  Schematic diagram of the linear Stark effect. The individual Stark states are labeled $|n, n_1, n_2, m\rangle$. (Taken from Jeys$^3$).
energy state of the \( n \) manifold will cross the lowest energy state of the \( n+1 \) manifold. As a general rule, this occurs at an electric field given by \( F = 1/(3n^5) \).\(^3\) The Stark structure of the atom after the manifolds cross becomes rather complicated.

From Figure 1.2, it is clear that the application of a strong electric field will strongly affect the interaction of Rydberg atoms with surfaces. Some Stark states will be more oriented towards the surface, and some will be more oriented away from the surface. Those states with the greatest electron probability densities near the surface will ionize when the core ion is furthest away. Thus, each Stark state for a given principal quantum number \( n \) will ionize at a different distance from the surface. For xenon, the Stark structure becomes intractable after the manifolds cross.\(^5\) Therefore, the experiment requires balancing the needs of keeping a high enough electric field to pull the ion away from the surface, but low enough to keep the Stark manifolds of adjacent \( n \) states from disturbing the energy level structure in ways that aren’t well understood.
Chapter 2: Theory

A detailed understanding of the current theory regarding Rydberg atom interactions with surfaces is a necessary guide in the design and development of the experiment. Until recently, a satisfactory theory of resonant electron tunneling processes for excited atoms near surfaces hadn't been developed. An early theory developed by Chaplik\(^6\) dealt only with atom-surface distances that were much larger than the classical size of the Rydberg atom. Later experiments with wire meshes\(^7\) implied an interaction distance of approximately \(4.5a_0n^2\), which, though larger than the classical orbit of the valence electron, is not much larger. A later attempt by Wille\(^8\) also proved unsatisfactory because he neglected the perturbation of the excited electron's orbit due to its own image charge interaction with the surface. While this work provided qualitatively interesting results, it could not provide quantitative results that could be verified experimentally.

More recently, a much more detailed treatment of the subject was provided by P. Nordlander and F.B. Dunning\(^9,10,11\) and by Kurpick \textit{et al.}\(^12\). In these treatments, both the effect of the applied electric field (necessary for detecting the resulting ions) and of the image charges of the electron and core ion in the surface are included. I will provide only a brief overview of the theory here with particular emphasis on the results.

In short, because the effect of the image potentials and the surface potential can be quite large for the atom-surface separation at which resonance ionization occurs, a perturbative approach is not appropriate. To properly describe the interaction, one must solve the Schrödinger equation for the potential (cylindrical coordinates):

\[ V_{\text{eff}}(\rho, z; Z) = V_0^S(z) + \Delta V_A(\rho, z; Z) + V_E(z), \quad (2.1) \]
where the first term on the right describes the interaction of the surface with the electron, the second term involves the effect of the proton, and the last term is the potential due to the externally applied electric field. Using resonance boundary conditions, one can deduce the complex energies of the eigenstates, $\varepsilon = \varepsilon_R - i\Gamma/2$, where $\varepsilon_R$ is the energy of the state and $\Gamma$ is the width of the state.

Figure 2.1 shows the results of the calculations for the ten $m=0$ Stark states of $n=10$ Hydrogen. The energies and widths of the states are plotted as functions of the atom-surface separation $Z$ for several values of electric field. One can immediately see that some of the Stark states are much more affected by the presence of the surface than others. From figure 1.3, we see that the energies of the Stark states are spread out more as the electric field is increased. We also see this in figure 2.1, where the energy shifts of the states are larger for increasing values of the electric field. Of even more importance from an experimental point of view is the effect of surface proximity on the width of the states. Since the width of the state is related to its lifetime, the width will determine the ionization rate as a function of atom-surface separation. The plot clearly shows that the width of the state can increase by several orders of magnitude over a relatively small change in $Z$. Also, the widths of the different Stark states vary greatly for a given atom-surface separation.

The reasons for these results become apparent when one looks at the actual shapes of the Stark state wavefunctions far from the surface and near to the surface. Figure 2.2 shows contour plots of the ten $m=0$, $n=10$ hydrogenic Stark state wavefunctions for an electric field of about 5kV/cm and two values of $Z$: one far from the surface and one near the surface. For large $Z$, one can see that some stark states are strongly oriented
Figure 2.1 Energy levels (left) and widths (right) calculated by Nordlander and Dunning\(^1\) for the \(m = 0, n = 10\) states of Hydrogen as a function of the atom-surface separation \(Z\) for three different electric field strengths. The dashed lines correspond to the same states shown in figure 2.2.
Figure 2.2 Contour plots of the probability density for the ten $m = 0, n = 10$ states of hydrogen for two distances from the surface. The upper plot is for $Z \approx 5,000 \text{Å}$ and the lower plot is for $Z \approx 150 \text{Å}$. The electric field is $E \approx 5 \text{kV/cm}$. (Taken from Nordlander and Dunning $^{10}$).
towards the surface, and some are strongly oriented away from the surface. The state oriented toward the surface mirrors in shape the state oriented away from the surface. The lower plot shows the same states when \( Z = 150 \text{Å} \). The Stark states oriented toward the surface are significantly perturbed, and, as expected, these states correspond to those in figure 2.1 with the largest width. Those states oriented away from the surface are little affected by the nearby surface.

For a beam of Rydberg atoms incident on a surface at near grazing incidence, the ionization distance will depend on the particular Stark state populated. The ionization rate is the rate of change of this population \( n_\sigma(t) \) with lifetime given by the formula

\[
\frac{dn_\sigma(t)}{dt} = -\Gamma_\sigma(t)n_\sigma(t) \quad (2.3)
\]

which can be solved by integration. The \( Z \) dependence of the positive ion production for a particular state \( \sigma \) with a perpendicular velocity \( v_\perp \) is then given by

\[
\frac{dn_\sigma(Z)}{dZ} = n_\sigma(Z) \frac{\Gamma_\sigma(z)}{v_\perp} \quad (2.4)
\]

which can be summed over all the states populated to give the total ion production as a function of atom-surface separation. A plot of this for the \( m=0, n=10 \) states of Hydrogen is given in the top portion of Figure 2.3. As the electric field is increased, the ions are produced ever farther from the surface.

If ions are to be detected, an electric field strong enough to overcome the Coulomb attraction of the ion with its image charge must be applied.
Figure 2.3  Top picture: Ion production as a function of atom surface separation $Z$. Bottom picture: Minimum electric field necessary for retrieving an ion at a distance of $Z$ from the surface. The solid line is for $T_\perp = 0\meV$, the dashed for $T_\perp = 1\meV$, and the dotted for $T_\perp = 10\meV$. (Taken from Nordlander and Dunning $^{10}$).
perpendicular to the surface. The necessary electric field can be derived by
considering a singly charged ion approaching a conducting surface with a
perpendicular kinetic energy \( T_\perp = \frac{mv_\perp^2}{2} \). Considering only motion along
the z-axis, the equation of motion for the ion is given by

\[
mv \frac{dv}{dz} = -eE + \frac{e^2}{4\pi \varepsilon_0 (2z)^2}
\]  \hspace{1cm} (2.5)

where \( z \) is measured from the surface and the ion-surface separation would
be \( -Z \). Integrating (2.5) once yields

\[
\frac{1}{2} m (z^2 - \nu_\perp^2) = -eE(z + Z) - \frac{e^2}{8\pi \varepsilon_0} \left( \frac{1}{z} + \frac{1}{Z} \right)
\]  \hspace{1cm} (2.6)

By setting \( \dot{z} = 0 \) one can derive the distance from the surface at which the
ion turns around and moves away from the surface, and a quadratic
equation for \( z \) results. The radical in the quadratic equation must be
positive, and this gives the solution from which one can derive the critical
electric field necessary for pulling the ion away from the surface. In atomic
units, this critical electric field is

\[
E_c(Z, T_\perp) = \left[ \frac{1}{2Z} + \sqrt{\frac{T_\perp}{Z}} \right]^2
\]  \hspace{1cm} (2.7)

The bottom of figure 2.3 shows the critical electric field as a function of
atom-surface separation for several values of perpendicular kinetic energy.
The plot clearly shows that the initial perpendicular component of ion
velocity can dramatically affect the electric field necessary for detecting the
ions. Keeping this in mind, the experiment must be designed to allow the
shallowest approach possible to minimize \( \nu_\perp \).
One can see from the theory outlined in this chapter that the experiment must delicately balance the need for an electric field strong enough to detect the resulting ions but low enough to avoid field ionization. The quantitative results derived from this theory pertain to hydrogenic Rydberg atoms. The experiment will be performed with xenon Rydberg atoms, which have Stark structure more complicated than hydrogen. Also, it will be difficult to realize electric fields greater than 4x10^6 a.u. (~20kV/cm) in the current apparatus. This limitation may restrict the experiment to principal quantum numbers greater than 10, but higher n adds more complexity to the theory. Certainly, there is still room for improvement on the theory as it relates to this particular experiment.
Chapter 3: Experiment

A. General Overview

Previous experiments on Rydberg-surface interactions proved for the most part unsuccessful. An early experiment by Kupriyanov\textsuperscript{13} measuring transmission of excited atoms through a grid simply noted that highly excited atoms are ionized by a nearby surface. He noted that this could be used as a means of detecting highly excited atoms, but little else was done with the results. Much later, Fabre \textit{et al.}\textsuperscript{7} performed a similar experiment with a beam of sodium Rydberg atoms passing through an array of slits. They noted that the transmission of Rydberg atoms through the slits using a "hard sphere" model implied an atomic size of $r=4.5a_0n^2$ for the excited atoms. This result is larger than that expected from a simple image model calculation, so the experimenters attributed their unexpected results to local electric fields due to impurities on the surface of the slits.

Indeed, later experiments by McCown \textit{et al.}\textsuperscript{14} and Gray \textit{et al.}\textsuperscript{15} confirmed Fabre's guess. By measuring the transmission of lithium Rydberg atoms through a gold mesh as a function of time, McCown \textit{et al.} observed the changes in transmission due to contamination of the mesh by the Li beam and then by absorption of background H$_2$O by the Li adsorbed onto the mesh. Model calculations of the process confirmed their results. This same surface contamination prevented Gray \textit{et al.} from determining the ionization distance of potassium Rydberg atoms from a gold surface. In this latter experiment a beam of K(nd) Rydberg atoms was directed at a flat gold mirror at a 2° angle of incidence with an electric field applied perpendicularly away from the surface that was used to accelerate products to a position sensitive detector. During the experiment, a layer of potassium was deposited on the surface even though the surface was
maintained at a temperature of 80°C, and this layer reacted with background 
H₂O to form a surface with unpredictable dipole electric fields. These 
spurious fields caused K⁺ ions produced by resonance ionization to be 
reflected from the surface with almost unit probability. Because of this, 
ionization distances could not be determined.

The present experiment seeks to overcome the limitations of the 
ellier experiments by using noble gas Rydberg atoms and performing the 
experiment in an ultra-high vacuum (UHV) chamber. Inert noble gases are 
excellent candidates for surface probes because they do not chemically alter 
the surface under study. To reduce surface contamination by background 
gases, the experiment is performed in a UHV chamber. With the low 
pressures (~1x10⁻¹⁰ torr after 3 days of baking) attainable in the interaction 
chamber (see figure 3.1), surface cleanliness can be maintained over the 
course of several hours.

Because the transition from the ground state to a Rydberg state for 
noble gases requires vacuum UV for single photon excitation, a two step 
excitation scheme is used. The atoms are first excited to a metastable state 
in a dc discharge and then to a Rydberg state by a dye laser. Xenon was 
chosen for this experiment for two reasons: due to current laser technology 
its Rydberg states are more easily accessible, and some literature already 
exists for xenon Rydberg atoms.

A schematic diagram of the apparatus is shown in figure 3.1. Xenon 
gas is excited to the metastable state in a dc discharge and collimated into a 
beam in the source chamber. The beam passes through an intermediate 
chamber containing deflection plates for removing ions from the beam and a 
gas cell for analyzing the effect of photons created in the discharge. The 
beam is collimated by a circular aperture separating the intermediate 
chamber from the interaction chamber. In this chamber, the beam is
Figure 3.1  Schematic diagram of the experiment. The Xe metastable beam is created in a DC discharge in the source chamber and travels to the interaction chamber where it is excited to a Rydberg state by an Coherent 899-21 dye laser (not shown). The MIR can be used for general Rydberg studies and Rydberg-surface interaction experiments.
further collimated before entering the main interaction region (MIR), where both Rydberg excitation and Rydberg-surface interactions are studied. The beam can also be directed into a Stern-Gerlach analyzer for analyzing the purity of the metastable beam.

B. Metastable Atom Source

The metastable beam source is shown schematically in figure 3.2. A detailed description of different types of metastable sources and the theory describing the particular metastable source being used is described in detail elsewhere;\textsuperscript{16} only a brief description of the source will be provided here.

Xenon gas (99.995\% pure) is admitted into an alumina tube inserted into the vacuum chamber through an ultratorr seal. The gas undergoes supersonic expansion into vacuum through a \(~0.005\)” hole laser drilled in the end of the alumina tube. The vacuum chamber is pumped by a 6” Varian diffusion pump with an estimated pumping speed of 900L/s. With a water baffle between the diffusion pump and the chamber, a base pressure of \(~2\times10^{-7}\) Torr is reached. During source operation, a pressure of \(~9\times10^{-5}\) is maintained in the chamber. The expanding xenon gas is collimated by passing through a 0.025” hole in an aluminum skimmer placed down stream. The position of the alumina tube relative to the skimmer can be varied by means of three orthogonal micrometer translation stages. The stages are connected to a welded bellows holding the alumina tube.

A 0.125” tungsten rod is inserted down the length of the alumina tube. By applying a voltage between the rod and the grounded skimmer, a discharge is struck through the expanding gas. A 61k\(\Omega\) load resistor is placed between the rod and the high voltage supply to limit the current. The rod is biased negative to accelerate any positive ions created in the discharge toward the alumina tube and not down the beam line. Optimum running
Figure 3.2 Schematic diagram of metastable source chamber and biasing electronics. The initiating sparker is used to pulse the cathode to a higher voltage than normal operating conditions to initiate the discharge.
conditions were achieved for a source voltage of 1150V and a tube-to-skimmer distance of ~6mm. Under these running conditions the current through the discharge is ~11.5mA. Thus, the voltage between the cathode (tungsten rod) and the skimmer is ~450V. After ~1hr of running, the discharge heats the skimmer to a stable temperature of ~40°C. Just beyond the skimmer (see figure 3.1) two biased deflection plates 2cm apart are positioned to remove any negative ions or long lived Rydberg states created in the discharge. One plate is grounded, and the other is kept at +300V.

The discharge produces a broad range of excited states in the xenon beam, most of which decay back to the ground state. However, approximately one in 10⁶ atoms in the beam remains in one of two relatively long lived metastable states. The lifetimes of the 3P₂ and the 3P₀ metastable states of xenon are 43s and 13ms, respectively.¹⁷ Calculations show that the velocity of the metastables in the xenon beam are centered around ~336m/s. Since the distance from the source to the interaction region is 74cm, the fraction of metastable atoms that survive passage to the MIR in the beam are 100% and 83%, respectively.

C. Characterization of Source Performance

Several methods were used to analyze the performance of the source. One method involved placing a stainless steel plate at the location of the main interaction region and measuring the current of secondary electrons ejected from it by the metastable beam. This current can be related to the metastable atom flux if the secondary electron ejection coefficient, γₑ, is known.¹⁸ If a fraction of the current, β, is due to energetic photons, the flux of metastable atoms incident on the stainless steel surface can be estimated from
\[ \Phi = (1 - \beta) \frac{I_{ss}}{e \gamma_e A} \]  \hspace{1cm} (3.1)

where \( I_{ss} \) is the current measured off the stainless steel plate, \( e \) is the electron charge, and \( A \) is the area of the beam at the target. A \( 0.20'' \) (0.10'') diameter aperture between the intermediate and interaction chamber defines the \( \sim 38 \text{mm}^2 \) \( \sim 9.1 \text{mm}^2 \) beam size at the target. For reasons that will be discussed shortly, the photon contamination was estimated to be \( \sim 10\% \) for Ar and \( \sim 15\% \) for Xe.

Initially, the source was characterized using argon. The current off of the stainless steel sample was measured to be 1.1nA. Using \( \gamma_e = 0.1 \) (good to a factor of \( \sim 2 \) only), the flux in the metastable beam is estimated to be \( \sim 1.6 \times 10^{11} \) Ar* atoms s\(^{-1}\) cm\(^{-2}\) at the MIR. Similar current measurements made for the Xe* beam with a 0.10'' diameter aperture yielded 0.0022nA. With \( \gamma_e = 0.02 \) for Xe\((^3P_0)\) on stainless steel, the estimated Xe metastable atom flux is \( \sim 6.5 \times 10^9 \) Xe* s\(^{-1}\) cm\(^{-2}\). Some current measurements were also made on silicon for Ar and Xe with the 0.20'' aperture giving 0.7nA and 0.02nA, respectively. Secondary electron ejection coefficients for silicon are not known, but the ratio of Ar to Xe current off silicon is close to the ratio of Ar to Xe beam flux determined from stainless steel. Because the internal energy the \( ^3P_0 \) state of Xe is greater than that for the \( ^3P_2 \) state (9.4eV vs. 8.3eV), \( \gamma_e \) is expected to be lower for the \( ^3P_2 \) state. Therefore, the calculated Xe* flux is most likely an underestimate.

In discharge sources, photons are created that can have energies close to the ionization energy of the source gas. The shortest wavelengths of photons created in rare gas discharges are 80nm for argon and 119nm for xenon. Ultra-violet photons this energetic can cause secondary electron emission at surfaces, which complicates beam flux measurements, and
photons reflected into the channel electron multiplier (CEM) used for Rydberg atom detection can give spurious counts. Characterization of the photons in the beam is important for properly understanding source operation.

To determine the number flux of photons in the beam, a gas cell was placed in the intermediate chamber in order to remove the metastables from the beam. The gas cell is a 6.25” cylinder with an inlet tube and two 0.25” apertures through which the Xe* beam can enter and exit. The target gas flowing into the inlet tube is carefully controlled with a needle valve. The intermediate chamber is pumped by a 900L/s diffusion pump. The pressure in the intermediate chamber is \( \sim 6 \times 10^{-7} \) Torr when the metastable source is running. For the aperture sizes given, the pressure inside the gas cell is estimated to be \( \sim 300 \) times the pressure inside the chamber.\(^{16} \) When a metastable Ar atom collides with a ground state Ar atom in the background gas, metastability exchange with the background Ar atom can occur, thus removing the metastable Ar from the beam. For sufficiently high pressures (2x10\(^{-3} \) Torr) all of the Ar* are removed from the beam (the beam is “quenched”), leaving only neutral Ar and photons to enter the interaction chamber.

Allowing the quenched beam to strike a stainless steel plate as described previously, the current from the plate is only 5% of its previous value for similar running conditions. Assuming the secondary electron ejection coefficient is the same for UV photons as for metastable Ar, the metastable beam is 5% photons. Similar experiments were not performed for Xe.

A Stern-Gerlach (SG) device was also used to analyze the performance of the metastable beam. The SG device is described in detail elsewhere and will only briefly be summarized here.\(^{19} \) The beam is
collimated to a small size by two 0.1 mm slits placed 10 cm apart. The metastable beam then passes through an inhomogeneous magnetic field produced by an electromagnet and experiences a force \( F_z = \mu_B \cdot \nabla \bar{B} \) perpendicular to the propagation direction of the metastable beam. The magnetic moment of the metastable atom is proportional to its total angular momentum along the \( z \) axis, \( m_J \). States with different \( m_J \) are deflected from the main beam a different angles. The count rate of emerging atoms is measured with a CEM as a function of deflection angle. From the area under each peak, the number of atoms with different \( m_J \) in the beam can be determined.

The metastable states of both Ar and Xe are \(^3P_0\) \( (m_J=0) \) and \(^3P_2\) \( (m_J=0, \pm 1, \pm 2) \) states. Typical SG scans for both Ar and Xe are shown in Figure 3.3. The side peaks of both scans are due to the \( m_J = \pm 2 \) and \( \pm 1 \) components of the \(^3P_2\) state. Because the velocity of metastables in the beam is broadly spread around a most probable velocity, the \( m_J = \pm 2 \) and \( m_J = \pm 1 \) peaks are not resolved. Assuming the metastable states are statistically populated in the source, the area under each of the three peaks should be equal if energetic photons do not contribute to the central peak. Assuming that photons and metastables have an equal probability of being detected, the percentage of photons in the beam can be found by comparing the integrated area under the side peaks with that of the central peak. Any excess in the central peak points to photon contamination in the beam. Using this method, the typical photon contamination determined for Ar was \(~12\%\). Because any misalignment in the SG apparatus would tend to decrease the side peaks, this value is very likely an over estimate.

Upon first inspection, the SG scan for Xe seems to show a large photon contamination. However, the energy level structure of Xe (figure 3.4) shows a large difference (1.13 eV) between the internal energies of the
Figure 3.3  Stern-Gerlach profiles of metastable beam for (a) argon and (b) xenon. The CEM is positioned 50cm from the magnets.
metastable states; whereas the difference in metastable energies for Ar is only 0.17eV. If the detection efficiency of a metastable atom depends upon its energy above the ground state, one would expect the central peak of the Xe scan to be larger than the side peaks because the $^3P_0$ state is higher in internal energy than the $^3P_2$ state. Though we can gain little quantitative results from the scan, we can see that the Xe beam does contain Xe metastables.

These tests have shown that a small fraction of the metastable beam is composed of photons. Ultra-violet photons can be detected by the channel electron multipliers used in this experiment. The present experiment must therefore be designed to discriminate against such photons.

**D. Rydberg State Excitation**

The excited energy levels of noble gases heavier than neon are best described by a $j_cl$ coupling scheme, where the total angular momentum of the core, $j_c$, is strongly coupled to the orbital angular momentum of the excited electron, $l$. Because the valence electron is excited from a filled $p$ state, the core is left with $l_c = 1$ and $s_c = 1/2$. Therefore the core may have $j_c = 1/2$ or $3/2$. This leads to two widely spaced series corresponding to the two configurations of the core. The state of the core is denoted $^2P_{1/2}$ or $^2P_{3/2}$, where the 2 means that the core is a doublet. The excited state energy levels are given the notation $nl[K]J$, where $n$ is the principal quantum number of the excited electron, $l$ is the orbital angular momentum of the excited electron, $K$ is the vector sum of $j_c$ and $l$, and $J$ is the total angular momentum of the atom (core plus excited electron). The orbital angular momentum is labeled $l$ for the excited states with the lower energy core, $^2P_{3/2}$, and $l'$ for those with the higher energy core, $^2P_{1/2}$. Often, the state of
Figure 3.4 Partial term diagram of xenon energy levels. Arrows indicate transitions used in experiment.
the core, $5p^5(^2P_{3/2, 1/2})$, is redundantly given before the state of the excited electron.

Figure 3.4 shows the energy levels of Xe, and the arrows indicate single photon excitation from the $^3P_0$ state metastable state to two different Rydberg series:

$$6s'[1/2]_0 \rightarrow np[1/2]_1$$
$$\rightarrow nf[3/2]_1$$

(3.2)

Two ionization limits exist for noble gases because of the two possible states of the core. It is possible, however, to excite transitions from one series to another. These transitions have been observed and used to measure the energies of the Rydberg series for several values of $n$.\textsuperscript{221}

There are several interesting things to note about the excitation scheme above. First, the selection rules in $j, l$ coupling for electric-dipole transitions are $\Delta J = 0, \pm 1; \Delta K = 0, \pm 1; \Delta l = \pm 1$; and $\Delta j_c = 0$. It is apparent that, though the $j, l$ coupling scheme best describes the individual atomic states, it does not entirely account for the allowed transitions. Both of the transitions above violate the last selection rule. In addition, the $6s' \rightarrow nf$ transition involves a $\Delta l = 3$ jump. One would also expect to see transitions to the $np[3/2]_1$ states; however, these transitions are weak. In addition, for $n > 14$, the transitions to $np$ states become weaker than transitions to $nf$ states.

Knight and Wang\textsuperscript{21} have attributed the existence of the $6s' \rightarrow nf$ transition to configuration interaction of the $6s'[1/2]_0$ with the $5d[1/2]_0$ state. They calculate the metastable state to be 34% $5d[1/2]_0$. Therefore the apparent violation of the $\Delta l$ selection rule is better understood. Knight also attributes the decline in intensity of the $np$ transition to destructive interference between the $6s'-np$ and the $5d-np$ portions of the transition, though this explanation needs a detailed theoretical calculation for proof.
Regardless of the reasons, the $6s' \rightarrow n\ell$ transitions offer the best choice for single photon excitation of Xe Rydberg states. The wavelength range for exciting Xe from the $^3P_0$ metastable state to $n \geq 10$ through ionization is 487.1nm to 462.2nm. By contrast, the same range of excitation for the $6s' \rightarrow n\ell$ transitions of Kr are 375nm to 361nm. Lighter noble gases would require even more energetic photons for Rydberg excitation from the metastable state. Lasers are not currently available that have a broad tuning range in the mid-300nm region. Wavelengths in this region are commonly reached by doubling the output of a visible/IR dye laser or Ti:Sapphire laser, a method that usually provides low power. The wavelength tuning range for exciting Xe Rydberg states falls in the blue region of the visible spectrum. This can be reached by the output of a dye laser without doubling. It is primarily for this reason that Xe was chosen in the present study.

E. Laser System

Xe($n\ell$) Rydberg atoms are excited using the output of Coherent 899-21 actively stabilized, single mode ring dye laser with a linewidth of 500kHz. The dye laser is pumped by 4.1W from a Coherent Innova 200 Argon ion laser with optics for the long UV. Most of the power from the Argon ion is in the 351.1nm, 351.4nm, and the 363.8nm lines. The laser dyes used are Coumarin 47 (C47) and Coumarin 102 (C102). A power vs. wavelength curve for each dye is shown in figure 3.5. These two dyes together provide 30-70mW of power for transitions to the $9f$ state through ionization. Because its tuning range covers more of the excited states, C47 is the preferred dye.

A small fraction of the laser output (~2%) is split from the main beam and is used to measure the wavelength with a scanning Michelson
**Figure 3.5** Dye laser output power for Coumarin 47 and Coumarin 102 measured over a range of wavelengths. These two dyes allow excitation to $n = 10$ through ionization with at least 40mW of power.
interferometer (referred to as the "wavemeter"). The device operates by splitting the laser beam into two. When the two beams are recombined, they constructively or destructively interfere, depending on their difference in path lengths. By continuously varying the path length difference, a series of light and dark fringes are created where the two beams are recombined. Sending two lasers of different wavelengths into the interferometer allows the instrument to be used as a tool to measure wavelength if the wavelength of one of the lasers is accurately known. The number of fringes for each laser is counted while the path length difference is continuously varied, and the number of dye fringes is compared to the number of fringes for a stabilized HeNe. Due the difference in refractive indices of air at $\lambda=632\text{nm}$ (HeNe) and $\lambda=470\text{nm}$, the wavemeter introduces a systematic $+0.001\text{nm}$ error. This instrument allows accurate determination of wavelength for the dye laser to $\pm0.01\text{Å}$.

A 70cm focal length plano-convex lens is used to focus the dye laser beam to a spot size of $\sim300\mu\text{m}$ in the interaction region.

F. Main Interaction Region

The apparatus to be used in studies of Rydberg atom excitation and surface interactions is shown in figure 3.6. It is located inside of a bakable ultra-high vacuum (UHV) system. The chamber is pumped by a 400L/s Perkin Elmer ion pump as well as a 450L/s Balzers TPU turbo pump. For the present work, the chamber is not baked and the background pressure is $\sim1\times10^{-8}$. The laser enters the chamber through a flat silica view port. A gate valve separates the interaction chamber from the intermediate chamber.

The main interaction region can be operated in two modes. In the first, the Xe* beam travels between a flat sample and an extraction grid. This mode allows general studies of Rydberg excitation in an electric field.
Figure 3.6  The detector used for general Rydberg studies and the initial Rydberg-surface interaction experiment. The sample is a 1.00" diameter copper mirror. The grids are 70 lines per inch; the 3 grids give a transmission of 73%. The laser intersects the Xe* beam perpendicularly. A spacing of 0.200" (0.508 cm) between the mirror and extraction grid defines the interaction region.
In the second mode, the Xe* beam is directed onto the middle of the sample face at grazing incidence. This configuration allows surface studies using Xe Rydberg atoms.

The MIR (shown in figure 3.6) consists of a copper sample, an extraction grid, two grids to reduce field penetration, a CEM detector, and the stainless steel, grounded housing. The spacing between the sample holder and the MIR housing is 0.100”, as is the spacing between the grid holders and the MIR housing. There is a 0.200” (0.504cm) spacing between the sample face and the extraction grid; this space is referred to as the interaction region. A 0.015” aperture collimates the Xe* beam before entering the detector. This aperture can be biased to reduce secondary electron ejection from the metastable beam hitting the aperture plate. The Xe* beam exits the detector through a 0.300” hole. The laser intersects the Xe* beam perpendicularly and is captured by a beam dump at the bottom of the detector.

The sample is a 1.0” diameter un-coated copper mirror polished to a flatness of \(\lambda/40\) at 10.6\(\mu\)m and guaranteed surface roughness of <5nm. The Rydberg atoms are expected to ionize hundreds of angstroms from the surface, so this surface roughness should be adequate. Each grid is 1.0” in diameter and can be independently biased, as can the copper sample. With the small sample-to-grid spacing, the electric field should be uniform in the center of the interaction region.

The MIR is attached to a 360° rotatable flange whose rotation axis is a line directed perpendicularly out of the page (figure 3.6) through a point halfway between the centers of the extraction grid and the sample face. It is the point showing the path of the laser in the figure. By removing a 0.100” spacer, the rotation axis can be shifted to the center of the sample face. By rotating the interaction region 4.3°, the Xe* beam is made to pass
through the aperture again and into the center of the sample. By this means, the experiment can be switched from general Rydberg studies (Xe* beam through center of interaction region) to Rydberg-surface interaction studies (Xe* beam into sample at 4.3°).

Either electrons or ions are accelerated from the interaction region toward a wide-mouthed (~1") Dr. Sjuts® CEM. When the CEM is biased at +2400 volts from front (mouth) to back, it provides a gain of ~5x10^7. The charge pulse is then amplified by an Amptek A-101 charge sensitive preamplifier-discriminator, which provides a CMOS compatible, 5V, 300ns output pulse. The pulses are read by an Ortec Log/Lin ratemeter. The grid nearest the CEM is kept at the same voltage as the mouth to prevent oddly shaped field lines from adversely affecting charged particle detection. The extraction grid and middle grid are always kept at the same voltage.

In order to characterize the performance of the MIR, measurements were made of the background counts detected by the CEM when 1) the Xe* beam passed through the middle of the interaction region and 2) when the Xe* beam was incident on the surface at ~5°. The sample was held at ground and the grids were biased to detect either electrons or ions. When looking for electrons, the bias conditions were: \( V_{CEM_{back}} = +2500V \), \( V_{CEM_{front}} = +100V \), and the extraction grid bias is varied. When looking for ions, the bias conditions are: \( V_{CEM_{back}} = +400V \), \( V_{CEM_{front}} = -2000V \), and the extraction grid bias was varied. Plots of the results are shown in figures 3.7 and 3.8. The large number of electron counts observed when the Xe* beam strikes the surface results from secondary-electron ejection. However, when the grid is biased to repel electrons, the count rate drops precipitously leaving only the small photon background. Electrons are also detected when the Xe* beam is passed straight through the detector. This can be explained from UV photons in the Xe* beam reflecting from the
Figure 3.7 Electron counts for (a) metastable beam striking surface at \( \sim 4^\circ \) incidence and (b) metastable beam traveling straight through the interaction region.
Figure 3.8 Ion counts for the metastable beam hitting the surface at ~5°. The increase in counts is due to photo-ejected electrons from extraction grid being driven toward the CEM.
edges of the entrance aperture and causing photo-electron ejection from the grids. Once the extraction grid is biased more positive than the front of the CEM, the electrons cannot reach the CEM. The difference in base count rates for electron detection arises from non-specular reflections of UV photons in the Xe* beam from the sample face. The increase in ion counts at -2kV for the Xe* beam striking the surface (figure 3.8) is also caused by photo-electron ejection at the extraction grid. When the CEM bias is higher than the extraction grid bias, electrons created at the grid are driven toward the CEM. When the Xe* beam passes through the middle of the interaction region, a constant background of ~10Hz is observed when biased to detections. All counts for all situations go to zero when the Xe* beam is blocked from entering the interaction chamber.

Studies of Xe Rydberg excitation and of Rydberg-surface interactions will be done in ion-detection mode. Because of the low background counts in both modes of operation, a good signal to noise ratio is expected in the experiment.
Chapter 4: Results and Future Direction

The production of Xe(nf) Rydberg atoms with $n = 16-20$ was investigated by biasing the detector for counting ions and applying a small negative voltage ($\sim 1-2$ V) to the extraction grid. Large ion signals well above the background were observed when the laser ($\sim 60$ mW of power) was tuned to the correct wavelength. The ions detected resulted from blackbody photoionization of the Rydberg atoms, an effect studied by other researchers.\textsuperscript{22, 23, 24} The experimentally determined wavelengths for the transitions are given in table 4.1.

In order to find the Rydberg transitions with the laser, accurate measurements of energy for the high $nf$ states and for the $6s'$ metastable state of xenon are needed. The energy of the $6s'[1/2]_0$ state is $76196.79$ cm$^{-1}$. Several groups have measured the energies of the Xe(nf) Rydberg levels for $n=8$ through $n=73$.\textsuperscript{21, 25-27} Some overlap in $n$'s existed between different papers, and there were some discrepancies between energies for the same state that were outside of the quoted uncertainties. These variances could correspond to as much as $\sim 0.2\text{\AA}$ difference in wavelength. The maximum scan width of the dye laser is $\pm 15$ GHz, which corresponds to a wavelength scan of $\pm 0.05\text{\AA}$. Thus, an inaccurate determination of the wavelength necessary for exciting a transition introduces great difficulties in finding the state.

The energy of a particular $n$ state can be estimated from the Rydberg formula

$$E_n = E_0 - \frac{R}{(n - \delta l)^2}$$  \hspace{1cm} (4.1)

where $E_0 = 97,833.81$ cm$^{-1}$ is the first ionization potential, and $R = 109,736.86$ cm$^{-1}$ is the Rydberg constant for xenon. The energies of the $f$ states for $8 \leq n \leq 45$ from Moore,\textsuperscript{27} L'Huillier et al.,\textsuperscript{26} and Knight et al.\textsuperscript{21} were
<table>
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<tr>
<th>$n$</th>
<th>$\lambda$ (literature)</th>
<th>$\lambda$ (interpolated)</th>
<th>$\lambda$ (measured)</th>
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<tr>
<td>9</td>
<td>493.429 nm</td>
<td>493.420 nm</td>
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<tr>
<td>10</td>
<td>487.134 nm</td>
<td>487.134 nm</td>
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<tr>
<td>11</td>
<td>482.583 nm</td>
<td>482.590 nm</td>
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<td>12</td>
<td>479.191 nm</td>
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<td>469.565 nm</td>
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<tr>
<td>21</td>
<td>467.556 nm</td>
<td>467.576 nm</td>
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**Table 4.1** The wavelengths for the $6s'[1/2]_0 \rightarrow nf$ transitions relevant to the current experiment are listed. Wavelengths in the first column were determined from energies given in the literature ($n=9-11$ from Moore $^{27}$ and $n=12-21$ from L’Huillier, *et al* $^{26}$). The interpolated quantum defect (line in figure 4.1) was used to estimate the wavelengths in the second column. The last column are wavelengths measured in the current experiment.
Figure 4.1  The quantum defect for nf Rydberg states of xenon determined from the energies listed by Moore, L’Huillier et al., and Knight et al. The line is drawn to interpolate between the high and low n measurements and provides a good estimate of the actual energy levels (see text). The dark squares show the quantum defects determined from the experimentally measured transitions.
used to obtain the quantum defect, \( \delta \), for each \( n \). The results are plotted in figure 4.1. The Rydberg-surface interaction experiment is expected to be performed for \( 10 \leq n \leq 20 \). Because the quantum defect is expected to remain roughly constant over a wide range of \( n \), some of the energy values are suspect. Instead of using the energy values listed in the paper to determine the wavelength of the transition, a line was drawn to interpolate between the high and low \( n \) measurements, and this value of \( \delta \) was used to determine the energy of the state. Table 4.1 shows the \( 6s' \rightarrow n=9, ..., 21 \) transition wavelengths determined from quoted values in the literature, from the interpolated quantum defect, and from measurements in the present experiment. Some discrepancies between the quoted values and measured values are clear.

Because the linewidth of the laser (500kHz) is much narrower than the resolution of the wavemeter, the excitation peaks were detected by tuning the dye laser to the expected wavelength and manually scanning the frequency very slowly over several GHz. When doing this, it was noticed that several peaks separated by hundreds of MHz occurred near the expected transition. The frequency was then tuned to the largest peak for \( n = 20 \), and a 5GHz scan centered on this peak was made under computer control. The results are shown at the top of figure 4.2.

Several closely spaced peaks are obvious. This structure is not entirely unexpected because there are nine stable isotopes of xenon. The hyperfine splitting of a transition is affected by the interaction between the nonspherically symmetrical charge distribution in the nucleus and the nonspherically symmetrical electric field produced by the atomic electrons, and different isotopes of Xe would certainly have different charge distributions in the nucleus. The theory for the hyperfine splitting of the \( 6s'[1/2] \) to \( 20f \) Rydberg transition in Xe would be quite complicated, but
Figure 4.2  $n = 20$ ($\lambda = 468.14\text{nm}$) blackbody photo-ionization signal over a 5GHz scan for (a) 2V/cm electric field and (b) 20V/cm electric field.
this must certainly be the effect present. Similar isotopic structure was seen by Walhout et al.\textsuperscript{28} in using the $6s\left[\frac{3}{2}\right]_2 \rightarrow 6p\left[\frac{5}{2}\right]_3$ transition to trap metastable xenon. By tuning a narrow linewidth Ti:Sapphire laser, they selectively trapped different isotopes of Xe. The spacing between levels ranged from a hundred MHz to over a GHz. For this reason, it is believed that the structure seen in figure 4.2 is due to the presence of different isotopes of Xe in the beam.

Because excitation in an electric field is of interest to the Rydberg-surface interaction experiment, scans similar to the one described above were performed for various values of electric field in the interaction region. Plots for field values of 20 and 50 V/cm are shown in figures 4.2 and 4.3. Changes in the structure from 2 V/cm to 20 V/cm are clear. More peaks appear, and the entire collection of transitions seems to have shifted (the center frequency of the scan is identical). Even more change in structure is seen in the 50 V/cm scan. More importantly, the center frequency of the scan is shifted down by 7.5GHz. This Stark shift is expected to play a major role in the studies of Rydberg-surface interactions.

In order to determine the saturation power for the transition, the laser was tuned to the largest peak shown in figure 4.2. Several neutral densities filters were used to lower the laser power and the corresponding drop in ion signal was recorded. The results are shown in figure 4.4. Though fluctuations in ion signal caused the points to be somewhat erratic, the plot shows that 70% of the signal is retained at only 30% of the laser power. Thus, the laser provides adequate power for saturating the transition.

In conclusion, much progress has been made toward studying Rydberg atom-surface interactions. An apparatus has been constructed in which both gas phase and surface Rydberg studies are possible. It was shown that noise from vacuum UV photons in the metastable beam is low in
Figure 4.3  Blackbody photo-ionization spectrum for n=20 ($\lambda$=468.140nm) and E=50V/cm. The frequency offset is referenced from the zero frequency of the scans in figure 4.2.
Figure 4.4  Plot of fractional ion signal for \(n=20\) Rydberg atoms vs. fractional laser power. The curve is a simple exponential fit. This signal is clearly near saturation for maximum (~60mW) laser power.
both modes of operation. Two laser dyes have been characterized that provide broad tuning ranges over the transitions relevant to the experiment. Several species of Xe(nf) Rydberg atoms have been created. The blackbody photoionization signal was well above noise levels. Using the \( \sim 2 \mu s \) lifetime of Xe(20f) and the fact that blackbody radiation ionizes \( \sim 1 \) in 1000 \( n=20 \) Rydberg atoms,\(^{22}\) the peak count rates in figures 4.2 and 4.3 imply excitation rates of \( 100 \)’s of kHz. This should provide good signal to noise ratios in future experiments. Finally, the experiment has identified systematic errors in earlier work.

The structure seen in figures 4.2 and 4.3 suggest that much general work needs to be done on the behavior of Xe(nf) Rydberg atoms in an electric field. The experiment is currently undergoing minor modifications to perform pulsed experiments. By pulsing the laser and then applying a field ionization pulse after a fixed time, lifetimes of Rydberg states excited in varying electric fields can be studied. The lifetime studies will determine the distance from the surface at which the Rydberg atoms can be excited while still maintaining useful count rates for the experiment. An alternative to exciting in an electric field is to excite the Rydberg atoms in a field free region before entering the region of high electric field. Lifetime measurements will show the feasibility of this approach.

The experiment is currently designed to study resonance ionization of Xe Rydberg atoms near the surface of a flat copper mirror. With the current MIR design, there is no way to remove the sample from its housing under vacuum and clean and characterize the surface. However, because resonance ionization is expected to occur hundreds of angstroms from the surface, the overall effect shouldn’t be strongly affected by a thin layer of contaminants on the surface (such as carbon deposits or oxidized copper).
Once the current method has been shown to be an effective way of studying Rydberg-surface interactions, a number of exciting effects can be studied. The interaction chamber is equipped with several tools for modifying and characterizing surfaces. After resonance ionization has been studied on the copper mirror, similar experiments should be performed on clean, flat metal samples like gold or copper. These experiments will determine what role surface contaminants on the copper mirror played in the first study.

By depositing thin metal films on an insulating material, a variety of effects can be studied. The process of resonance ionization of Rydberg atoms is expected to occur hundreds of angstroms from the surface, but the capability exists to deposit layers of metal only several angstroms thick. Contrary to the bulk material, electrons in a thin layer of metal may behave much like a particle in a square well potential with discrete energy levels. The surface potential of thin films would be very different for thin films than for thick conductors. Resonance ionization of Rydberg atoms near surfaces would be very different for thin films. Thus, the effect of thickness of the thin film on Rydberg-surface interactions could be studied. As a further exploration, one could deposit an insulating layer on top of a thin conducting layer. The question of whether the electron can tunnel through the insulating layer to the conductor below presents an interesting problem.

Velocity selective excitation (VSE) is also possible in exciting the Xe metastable atoms to a Rydberg state. If the laser and the metastable beam are not perpendicular, the Rydberg transition becomes Doppler broadened. With the narrow line-width dye laser, it is possible to excite only metastable atoms in a small range of velocities to a Rydberg state. As shown in figure 2.3, the critical electric field depends strongly on the velocity of the atom.
By tuning the electric field close to the critical point for ion detection, it should be possible to turn the signal on and off with velocity selective excitation. In place of the CEM in the current experiment, a position sensitive detector (PSD) could be used to gain information on the arrival location of the ions. With VSE capabilities and a PSD, it could be possible to image the varying electronic potentials near the surface of an electronic device using resonance ionization of Rydberg atoms. In summary, resonance ionization of Rydberg atoms promises to be an exciting method for probing electron tunneling processes near a variety of surfaces.
References