Search for novel molecular species formed by cold Rydberg atoms in a magneto-optical trap

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A Search for Novel Molecular Species
Formed by Cold Rydberg Atoms in a Magneto-Optical Trap

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Abstract

This describes a search for novel ultra-long-range molecular resonances between rubidium Rydberg atoms in a magneto-optical trap (MOT). We use the technique of photoassociative spectroscopy by directly scanning the wavelength of our Rydberg excitation laser about a $5p_{3/2} - 90p_{3/2}$ transition in a weak electric field and by scanning mm-waves about a $39s_{1/2} - 39p_{3/2}$ transition. The results of both of these surveys are inconclusive.
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Chapter 1: Introduction

My goal in this experiment was to observe evidence of long-range molecular resonances involving Rydberg atoms in a magneto-optical trap (MOT). A Rydberg atom is an atom of high principal quantum number $n$; generally atoms with $n \geq 10$ may be considered Rydberg atoms. This experiment considers Rydberg atoms with a single highly excited valence electron, namely Rubidium from the alkali metal group. While the Bohr model for single-electron atoms is inherently flawed in its interpretation of atomic structure, it provides a useful demonstration of the properties of interest in the study of Rydberg atoms. According to the Bohr model, the orbital radius of an atom's valence electron is given by

$$r = \frac{4\pi \varepsilon_0 n^2 \hbar^2}{Ze^2 m_e} \propto n^2,$$

where $n$ is the principle quantum number, $\varepsilon_0$ is the permittivity of free space, $Z$ is the atomic number, $e$ is the electron charge, $m_e$ is the electron mass, and $\hbar = h/2\pi$, where
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\( h \) is Planck's constant. The binding energy of the electron, on the other hand, is given by

\[
E = \frac{-Z^2e^2m_e}{\left(4\pi\varepsilon_0\right)^2 2n^2h^2} \propto n^{-2}.
\]

Hence, a Rydberg state is characterized by the large orbital radius and relatively weak binding energy of its valence electron. The large orbital radius results in a large electric dipole moment which heavily influences the interactions between Rydberg atoms. Furthermore, Rydberg states demonstrate long radiative lifetimes relative to those of lower energy states. It is this essentially "extreme" nature of Rydberg atoms which makes them interesting test cases in atomic physics. Because they are so close to the ionization limit, stable Rydberg atoms do not occur naturally on earth. They can readily be produced in the laboratory, though, by means of collisional excitation, optical excitation, or other techniques. Note that the study of Rydberg atoms is not a topic of interest for purely atomic physics, but it also has practical applications in various other fields. For example, the long radiative lifetimes of Rydberg atoms enable them to persist in the near vacuum conditions of interstellar space, causing them to have relevance in a significant body of astrophysical study [1]. Also, the ability of molecules to possess Rydberg states has implications for both physical and organic chemistry [2]. It is my goal in this experiment to observe experimentally one type of extremely long range molecular interaction between pairs of rubidium Rydberg atoms.
The study of molecular resonances involving Rydberg atoms has become a topic of growing interest in recent years. As one would expect for any type of reactive atom, Rydberg atoms possess molecular potentials. These potentials are greatly influenced by the dipole-dipole, or van der Waals interaction, and they vary depending on the states of the atoms. As with ground state atoms, Rydberg atoms are capable of forming bound states with other atoms. The difference here is that since Rydberg atoms are near the ionization limit, these bound states are likely to be very shallow, making them nearly impossible to observe within a hot gas. For reasons that I will explain below, the ideal environment to observe molecules such as these is an ultracold gas such as the optically cooled contents of a MOT.

The key technique we use to observe molecular resonances between Rydberg atoms is photoassociative spectroscopy. Photoassociation is the process by which a pair of atoms is excited to a higher molecular energy state by the collective absorption of radiation during an atomic collision. Among its many uses, this technique has been proposed as a method to stitch ultra-cold, trapped atoms together into polar molecules to serve as a quantum memory [3]. Photoassociation is a well-known technique for the production of bound molecules, but in the case where the target atoms have initially been optically cooled, the technique can also be used for spectroscopy. After the molecular excitation occurs, there are several possible outcomes. The molecule either de-excites and disassociates at high energy, de-excites to a molecular ground state, or excites to a higher level and ionizes. Each of these cases results in the constituent atoms falling out of the trap, resulting in observable spectra.
room temperature atoms there is such a large spread in thermal velocity that, by virtue of the Doppler shift, there is a tremendous variation of the energy of incoming photons as perceived by the molecules, this is not the case for optically cooled and trapped atoms. Thus, hot atoms cannot produce spectra with sufficient resolution to observe any evidence of these resonances, but cold atoms which have kinetic energies in the range of $\sim$100$\mu$K can produce such spectra [4].

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{potential_curves.png}
\caption{Example molecular potential curves between Rydberg atoms. Two pairs of potentials are shown with asymptotic energies $E_1$ and $E_2$. In reality many attractive and repulsive potentials could share the same asymptotic energies. Two of these potentials exhibit an avoided crossing at inter-nuclear separation distance $a$ (inset). Avoided crossings such as these may occur at very large separation distances.}
\end{figure}

This technique also allows one to observe a class of very long range molecular resonances. At inter-atomic separations greater than those for molecular bound states there may be points where a lower energy repulsive potential coincides with a higher
energy attractive potential as shown in Fig. 1. At this range the Hamiltonian may become perturbed such that these two potential curves experience what is referred to as an avoided crossing. The attractive potential may then possess a shallow bound state in the region of the avoided crossing. Because these states occur at such long range, however, their energies will be very near the asymptotic values of the potential. Therefore, it would be extraordinarily difficult to excite hot atoms into these states, but optically cooled atoms could be excited with the requisite precision to attain them [4].

There is also a type of ultra-long range molecule called a macrodimer that exists between pairs of Rydberg atoms. These molecules are caused by the emergence of potential minima at extremely long range arising from the scaling of the dispersion coefficients for the molecular potential for certain pairs of Rydberg states [5]. There is indirect evidence for the existence of this type of molecule: the theoretical potentials were used to create a model for the time evolution of an energy transfer collision between Rydberg atoms for various values of the energy difference before and after the collision. This model showed excellent agreement with experiment [6].

Furthermore, a class of long range bound molecules has been predicted to exist between Rydberg atoms and ground state atoms as a result of Born-Oppenheimer potential curves, so called “trilobite molecules.” These molecules occur in two types, the first having low orbital angular momentum ($l$) values for the Rydberg atom's valence electron and the second having high orbital angular momentum values. In both cases many minima occur in the molecular potential due
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to the oscillating character of the Born-Oppenheimer curves as functions of the inter-atomic separation. Interestingly, the high $l$ type is predicted to have deep potential wells supporting multiple vibrational states. Furthermore, these states are predicted to have permanent strong electric dipole moments, a feature not present in any other homonuclear diatomic molecule. The ability to manipulate the polarizations of these molecules would provide a useful tool to further study long range molecular states.

These "trilobite-like" molecules show striking complexity in their electron probability densities, one such example is shown in Fig. 2. Evidence for the existence of these molecules has yet to be experimentally observed [7].

![Figure 2: A plot of electron probability density for a “trilobite molecule” (cylindrical coordinates). The location of the Rydberg ion is represented by a white dot. The ground state atom is located at the large double peak [7].]
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A group at the University of Connecticut at Storrs observed yet another novel effect in which Rydberg atoms showed evidence of molecular resonances at extremely long range. This group did photoassociative spectroscopy of cold rubidium Rydberg atoms in a MOT by directly exciting them from 5s ground states to np states where n ranged from $n = 50$ to $n = 90$. This excitation was done using high resolution UV laser. This group observed long range molecular resonances slightly to the low frequency side of the np + np resonance. Although similar to the predicted macrodimer states discussed above, the observed resonances did not correspond to the macrodimer models, nor did they correspond to the predicted trilobite states. Instead, they were found to correspond to the average energy of the $(n - 1)d + ns$ asymptotes. Note that both the transitions from the 5s ground state to the $(n - 1)d$ and the $ns$ states are forbidden by the angular momentum selection rules. They theorized that these resonances could be attributed to avoided crossings between the $(n - 1)d + ns$ molecular potentials and a highly attractive np + np potential. The strong dipole coupling between each of these states and the np state results in the $(n - 1)d + ns$ curve taking on the character of the np state. This allows the observed excitation to the $(n - 1)d + ns$ curve. This group also saw a similar feature corresponding to the $(n - 1)p + (n + 1)p$ potential curve which can be explained analogously but with a quadrupole coupling rather than a dipole coupling. Interestingly, these avoided crossings occur at 58000$a_0$ and 75000$a_0$ respectively where $a_0$ is the Bohr radius. Indeed, these are extraordinarily long range interactions, considering that the average nearest neighbor distance of ground state atoms at the temperature and pressure of the
MOT is approximately $23000a_0$ [8]. Our goal in this experiment was to observe molecular resonances similar to these in the MOT in our laboratory at Colby College.
Chapter 2: Experiment

Because the observation of spectroscopic features resulting from molecular resonances between Rydberg atoms requires high resolution, the rubidium atoms whose Rydberg states are observed in this experiment are first optically cooled and contained in a magneto-optical trap (MOT). They are then excited to Rydberg states using a frequency-doubled, dye-amplified diode laser system. The Rydberg state populations may then be further modified using mm-wave spectroscopy. Detection is done using pulsed field ionization.

Magneto-optical Trap

The MOT used in this experiment is designed to trap rubidium-85. The principle of optical cooling is based on the slight recoil momentum imparted to an atom when it absorbs an incident photon. An incident laser beam which is tuned slightly to the red of resonance with this atom’s absorption frequency will be more likely to induce an excitation when the atom’s velocity is anti-parallel to the direction
Figure 3: The trapping laser in the MOT excites from the $5s_{1/2} F = 3$ state to the $5p_{3/2} F = 4$ state. Because the hyperfine splitting is so small, the uncertainty principle ensures that a fraction of these atoms end up in the $F = 3$ state. Of these $F = 3$ state atoms, some de-excite back into the $5s_{1/2} F = 3$ state, and some de-excite into the $5s_{1/2} F = 2$ "dark state." It is the job of the re-pump laser to return these atoms to the trapping cycle.

of the beam, thus blue shifting the laser frequency in the atom's perspective. Six laser beams incident on the atom from different directions are sufficient to ensure optical cooling regardless of the atom's direction in three dimensional space. In practice, we trap using an external cavity diode laser (ECDL) which is locked at the desired frequency then amplified, frequency isolated, beam expanded, and split, passing through the trap along the three cardinal directions and reflecting back upon itself. As shown in Fig. 3, the frequency of this trapping laser is locked just short of 780 nm, inducing a transition from the $5s_{1/2} F = 3$ ground state of $^{85}$Rb to its $5p_{3/2} F = 4$ excited state ($F = J + I$ is the atom's total angular momentum where $J$ is the orbital angular momentum plus electron spin, and $I$ is the nuclear spin). Although most of the atoms de-excite back to the $5s_{1/2} F = 3$ state, a small fraction go to the $F = 2$ "dark
Due to the relatively large energy differences in the hyperfine structure between the $5s\,^1S_2 F = 2$ and $F = 3$ states as compared to the $5p\,^3P_2 F = 2$ and $F = 3$ states, these atoms will no longer be cooled by the trapping laser and are effectively taken out of the trap. Therefore it is necessary to use a second re-pump laser to excite the $5s\,^1S_2 F = 2$ state atoms back up to the $5p\,^3P_2 F = 3$ state. A sufficient fraction of these de-excite back to the $5s\,^1S_2 F = 3$ state that the trap is maintained [9, 10].

This is sufficient to cool the atoms in the low velocity tail of the Maxwell distribution, but additional measures are required to prevent them from diffusing out of the trapping region. This is achieved by using a pair of anti-Helmholtz coils to set up a quadrupole magnetic field within the trap. This field varies linearly in all directions from the trapping point where the magnetic field $B = 0$. Furthermore, the trapping laser beams are all circularly polarized in the appropriate direction such that the following occurs. Imagine the magnetic field increases linearly in one dimension through the $B = 0$ trapping point. The magnetic field causes the angular momentum states of the trapping excitation to be perturbed by the Zeeman effect such that on the high-$B$ side of the trapping point, states with negative values for the magnetic quantum number $m$ are lower in energy, and on the low-$B$ side, states with positive $m$ values are lower in energy. This is shown in Fig. 4(b). Recall that the trapping laser is detuned from resonance such that a stationary atom at the $B = 0$ point is not likely to experience an excitation. At the high-$B$ side, however, this atom would be more likely to absorb photons which excite it to the lower $m$ state than the higher $m$ state. The opposite would be the case on the low-$B$ side. Thus, the excitation laser incident on
Figure 4: One dimensional depiction of the MOT. (a) If no B field is applied, both polarizations of incident photons are equally likely to induce a transition that imparts momentum to an atom regardless of the atom’s spatial position. Note that the photon energy is slightly detuned from resonance (dashed line) so as to selectively excite atoms in motion (for optical cooling). (b) If a linearly varying B field is applied, the $m = \pm 1$ states are no longer degenerate. Negatively polarized photons incident from the right are more likely to induce transitions in atoms on the right side of the trap and vice versa. This effectively constitutes a restoring force that pushes the atoms towards the $B = 0$ trapping point at the center of the trap [9, 10].

the atoms from the high-B direction should be circularly polarized with the proper handedness that selects for low $m$ excitations. A beam of opposite handedness should be incident from the low-B direction. Therefore, if the handedness of all six circularly polarized beams is properly chosen, an effective potential is set up which forces the optically cooled atoms toward the $B = 0$ trapping point. This is the principle which allows the MOT to function [9, 10]. The MOT used in this experiment traps approximately $10^8$ atoms in a roughly 1 mm diameter blob within a vacuum chamber held at low pressure by a 20 L/s ion pump. The thermal velocities of these optically
cooled atoms are approximately 0.2 m/s which correspond to a temperature of about 100 μK.

It is necessary at this point to excite the cold, trapped 85Rb atoms to Rydberg states. While it is common in some laboratories to use continuous wave optical excitation schemes, this experiment makes use of a pulsed optical excitation mechanism for cost effectiveness and low maintenance requirements. Our primary means of Rydberg excitation is through a pulsed, frequency-doubled, dye-amplified diode laser system. We also make use of a pulsed mm-wave excitation system. I shall now describe each of these excitation schemes in detail.

**Pulsed Dye Laser**

The pulsed, frequency-doubled, dye-amplified diode laser system shown in Fig. 5 illuminates the trapped atoms with a beam of power level up to approximately 0.4 mW at around 480 nm. We used a range of excitation wavelengths in this experiment. I shall now describe how the system drives a transition from the $5p_{3/2}$ state to the $39s_{1/2}$ state in $^{85}$Rb, a transition that requires an excitation pulse of 480 nm (blue) light. The power for this pulsed laser is provided by a neodymium-doped yttrium aluminum garnet (Nd: YAG) laser. This 532 nm (green) pulsed beam is split into three parts, one having 30%, one having 10%, and another having 60% of the total beam power. These are injected into three circulating liquid dye cells, having concentrations 0.06 g/L, 0.06 g/L, and 0.12 g/L of dye respectively. The dye we use is LDS 925 dissolved in ethylene glycol and polypropylene carbonate. The high-power
Figure 5: Photograph of the pulsed, frequency-doubled, dye-amplified diode laser system.

Nd: YAG laser excites the dye to a wide range of states setting up, in particular, a population inversion corresponding to the excitation of interest. The wavelength of the laser is controlled by the seed beam, in this case a 960 nm infrared beam provided by an ECDL. For precision we use a commercially available ECDL, a Sacher Lasertechnik “Lion” Tunable Littman/Metcalf ECDL. This beam passes through the three dye cells, causing stimulated emission. The resulting high-power IR beam is frequency doubled in a K\text{NI}O\text{3} crystal to produce the desired 480 nm output wavelength. Finally, the beam passes through a Pellin-Broca prism to isolate this
wavelength and is sent into the trap for excitation of $^{85}$Rb to Rydberg states. The length of these pulses is 10 ns, corresponding to the pulse length of the amplifying Nd: YAG laser.

**Millimeter Wave Spectrometer**

In some experiments, we use mm-waves as a tool for additional Rydberg excitations. The mm-wave band includes light frequencies between 30 GHz and 300 GHz corresponding to a wavelength range extending from 1 mm to 10 mm, hence the name. These frequencies are well suited for excitation between Rydberg states in $^{85}$Rb. From the $39s_{1/2}$ state provided by the pulsed, frequency-doubled, dye-amplified diode laser system, we use pulsed mm-waves to excite to various other Rydberg states. This two-photon excitation scheme provides an economical way to achieve a wide range of excitations. At the heart of our mm-wave system is an HP83630 mm-wave source. This device may be set to output a wide range of frequencies and power levels at a specified pulse length and delay. To transmit these waves into the trap, we use a waveguide with two frequency multipliers and a horn (a waveguide with rectangular cross section of linearly increasing dimensions). The waves emitted from the horn have been isolated at the desired frequency which is four times the frequency output by the HP83630. They are reflected by an off-axis paraboloid reflector which directs them through a window in the vacuum chamber and into the trapped atoms as shown in Fig. 6.
Figure 6: Photograph of the apparatus which transfers mm-waves generated by the HP83630 to the MOT vacuum chamber.

Detector

Our primary means of detecting relative populations of Rydberg atoms species within the trap is with field ionization. This operates on the principle that the electric field required to ionize neutral Rydberg atoms decreases with increasing \( n \). The Rydberg atom’s coulomb potential is perturbed by the electric field as is shown in the Fig. 7. Thus, as the strength of this ionization field is ramped, ever more deeply bound Rydberg states ionize, producing an electron and an \(^{85}\)Rb ion. We apply this field ionization pulse across two high-transparency mesh plates within the vacuum chamber. A micro channel plate (MCP) detector, which operates similarly to a
Figure 7: (a) The atomic Coulomb potential showing three energy states. (b) When a uniform electric field is applied, the Coulomb potential becomes perturbed. The topmost Rydberg state in (a) is no longer a bound state in (b). Atoms in this state will ionize and their positive ions will accelerate to the right when the field is applied.

A photomultiplier tube, is situated on one end of the chamber. Since the distance from the trapping point to the MCP is only a few centimeters, the electrons (or ions, depending on the field orientation) from a particular Rydberg state are accelerated towards and detected by the MCP almost immediately upon their forming a plasma.

Since the energy needed to field ionize is the binding energy of the Rydberg state, the time delay between the application of the field ionization pulse and the detection of electrons by the MCP is an indicator of the atom’s Rydberg state. This results in a time-of-flight spectrum of the trap’s Rydberg state populations. In practice, this is mostly useful to determine relative populations of states, but if the types of states present in the trap are known (as is often the case), then the time-of-flight spectrum is a powerful observational tool. Of course, this detection mechanism also eliminates from the trap the Rydberg populations of interest, but since the excitation methods we
use are pulsed rather than continuous wave, the trap can be quickly returned to its original condition, and many shots can be taken per second. This detection scheme is also used to take spectra as some external parameter, such as laser wavelength or mm-wave frequency, is swept. In this case a boxcar averager is used to integrate the signal from the time-of-flight spectra corresponding to a particular Rydberg state or group of states. The external parameter is then incremented over a specified number of shots, producing a spectrum for that parameter.

**Experiments**

We made use of two major approaches to search for these long range molecules in our laboratory. The first approach was to take spectra by directly scanning the frequency of the pulsed, dye-amplified Rydberg excitation laser. The second approach was to first excite to a Rydberg state using the pulsed laser, then perform mm-wave spectroscopy. I shall now discuss each of these approaches in detail.

**Pulsed Laser Wavelength Scan**

The first approach is a straightforward two photon excitation involving the $5s$ to $5p$ transition provided by the trapping laser and the $5p$ to Rydberg state transition provided by the pulsed dye laser. For this experiment we chose to excite to the vicinity of a $90p$ state corresponding to a laser wavelength of 958.7787 nm. Because
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this is not an allowed transition, we also applied a constant electric field across the trapping region to perturb the Hamiltonian. Control of the apparatus and data acquisition was done using a LabView program that I modified from Mao Zheng’s frequency scanning data acquisition program [11]. This program interfaces with a Stanford Research Systems SR245. The SR245 outputs a DC voltage to the Sacher laser controller. This voltage is applied to the piezoelectric crystal inside the ECDL seed beam for the pulsed dye laser which controls the frequency of output beam. Spectral data was obtained using pulsed field ionization as described above. The SR245 also reads the data. One channel is the spectral data from the boxcar averager, and the other channel is from an etalon which calibrates the laser frequency over the scan. The SR245 relays this information to the computer which assembles it into spectra. The program can run a specified number of shots over a specified voltage range to be applied to the ECDL’s piezoelectric crystal. In this experiment we incremented over a 10 V range corresponding to a frequency range of approximately 7.5 GHz.

Millimeter Wave Spectroscopy

The second approach we used was to take mm-wave spectra of Rydberg atoms. As described above we first used the pulsed dye laser to excite a transition from the trapping laser’s 5p state to a 39s state. We then used the mm-wave apparatus to inject waves at and around 68.376 GHz, causing an excitation from 39s to 39p. No external electric field was applied. Data were taken for both the electron field ionization signal
Experiment

and the electron signal from any plasma that formed as a result of attractive forces between Rydberg atoms that led to ionization. We used Mao Zheng’s frequency scan program [11] to acquire data in all of the mm-wave experiments.

In the next experiment we again excited to around the 39p state, but in this test we integrated over the pulsed field ionization signal rather than the plasma signals. The number of atoms experiencing this transition depends on the power of the mm-wave input. Using the time-of-flight spectra we can determine the ratio of these two populations within the trap. In this experiment there was a roughly equal ratio of 39s to 39p state atoms in the trap. The power of the pulsed dye laser was at 0.8 mW, and this time we observed only the electron field ionization signal. Data were taken for mm-wave frequencies between 67.2 GHz and 69.6 GHz. In this test we also observed the time evolution of the system by taking data for various delay times between the mm-wave pulse and the field ionization pulse. These delay times were 0 μs, 10 μs, and 20 μs.

Lastly, we repeated this experiment but with an adjustment in the mm-wave power so that there was a larger percentage of 39p state Rydberg atoms. In this test it was expedient to narrow the scan range. We scanned around the 39p absorption from 68.0 GHz to 68.8 GHz.
Chapter 3: Results and Discussion

**Pulsed Laser Wavelength Scan**

![Graph showing spectra for laser wavelength scan around the 5p to 90p transition with various voltages applied across the trapping region. No obvious evidence for molecular resonances is present.]

**Figure 8:** Spectra for laser wavelength scan around the 5p to 90p transition with various voltages applied across the trapping region. No obvious evidence for molecular resonances is present.
Results and Discussion

The results for the direct laser frequency scan are shown in Fig. 8 above. Because this excitation corresponds to a $5p$ to $90p$ transition which is forbidden, there is no prominent single peak. We see three resonances when no electric field is applied which most likely correspond to $d$ state transitions. The application of an electric field gives rise to Stark structure as can clearly be seen in the spectrum at 2.0 V. We do not, however, see any structure that could be obviously indicative of molecular resonances. Such structure either is not present, or is lost in the high frequency noise.

Millimeter Wave Spectra

We turn our attention then to the results of the mm-wave experiments. The results of the plasma signal study are shown in Fig. 9. This scan shows a significant broadening to the low frequency side of the $39s_{1/2}$ to $39p_{3/2}$ resonance. This is expected since we are looking at the plasma signal. The dipole interaction between Rydberg atoms includes both attractive and repulsive potentials. When a pair of Rydberg atoms is in an attractive state they will collide and ionize, producing a plasma. By looking at the plasma signal, we are selectively observing the signal from Rydberg atoms which have some likelihood of being at closer inter-atomic separations, and hence lower energy, than the asymptotic energy of the transition. This is illustrated in Fig. 10(a). Fig. 10(b) shows the type of feature that we would hope to see as an indicator of a molecular resonance similar to those observed by the group at the University of Connecticut at Storrs. We do not, however, see any such features within the noise limits of the experiment.
Results and Discussion

Figure 9: Millimeter wave spectra for the $39s_{1/2}$ to $39p_{3/2}$ transition: (a) the electron signal and (b) the ion signal of the plasma formed by Rydberg atom collisions along an attractive potential.
Results and Discussion

Figure 10: (a) Diagram of excitation to an attractive potential and the resulting spectra. The peak will be highest at the asymptotic energy of the potential curve but will show a broadening to the low frequency side as a result of lower energy transitions. (b) How the data might indicate a long range molecular resonance due to an avoided crossing on the same potential. In practice the spectra might instead show a dip at the avoided crossing rather than a satellite peak.

The next set of results is from the mm-wave experiment field ionization signal, and is shown in Fig. 11. Here we see an array of interesting features surrounding the $39s_{1/2}$ to $39p_{3/2}$ transition. Most prominent is the peak on the high frequency side of the transition. This corresponds to the two-photon $39s_{1/2}$ to $40s_{1/2}$ transition and not a molecular resonance. The origin of the other features is still in question. When a delay is added between the mm-wave pulse and the field ionization pulse, we see additional structure on both the high frequency and low frequency sides of the primary resonance. These features cannot be attributed to noise. In subsequent tests, however, we were unable to reproduce these data. A possible explanation is that having the mm-waves trigger before the Rydberg laser pulse was finished caused an unexpected interaction resulting in these features, but this is unconfirmed as of yet.
Figure 11: Millimeter wave spectra for the $39s_{1/2}$ to $39p_{3/2}$ transition: the electron field ionization signal with various time delays between the mm-wave and field ionization pulses.

Finally, Fig. 12 shows the results of this same test after having adjusted the mm-wave power to increase the fraction of $39p$ to $39s$ states. We find that the mysterious interference has disappeared, and instead we are left with a well defined $39s_{1/2}$ to $39p_{3/2}$ transition peak. We again see the $39s_{1/2}$ to $40s_{1/2}$ two-photon resonance to the high frequency side of this peak. No other structure is visible.
Figure 12: Same as in Fig. 11 except that the mm-wave power has been increased. The scan range has been narrowed around the $39s_{1/2}$ to $39p_{3/2}$ transition peak. A peak for the two-photon $39s_{1/2}$ to $40s_{1/2}$ excitation is clearly visible.
Chapter 4: Conclusion

We were unable to observe with certainty any evidence for long range molecular resonances within the MOT by photoassociation. Both of our approaches, the laser wavelength scan and the mm-wave scan, were inconclusive. It seems that our dye-amplified laser scan lacked the precision of the UV spectra used successfully in the original experiment [8], and it is still unclear whether or not analogous results can be obtained from mm-wave spectroscopy.

In future work it may still be possible to observe signs of the so called "trilobite molecules." It has been suggested that one could excite atoms into these high $l$ configurations using a three-photon transition. The 787.0 nm and 479.4 nm wavelengths which give rise to the $5s + 30d$ state are attainable using our trapping and pulsed dye lasers. The final microwave excitation to $5s + 30fgh$ states should be accessible using our mm-wave apparatus with additional frequency multiplication when a weak electric field is applied [7]. This research direction is well worth looking
Conclusion

into, but work must first be done to ensure that our mm-wave spectrometer is sufficiently precise to make the observation.
References


