Sympathetic Cooling of Molecules Using Laser-Cooled Atoms

Maya Lewin-Berlin

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Abstract

Sympathetic cooling of molecules using laser-cooled neutral atoms is a new technique being developed in the Hudson Lab at UCLA that may be able to cool clouds of molecular ions to their ground energy states. Molecular ions are trapped in an oscillating electric field inside a magneto-optical trap containing a cloud of ultracold neutral atoms. The collisions between the molecules and the ultracold atoms lead to significant cooling in the molecules. The development of efficient diagnostic techniques allows researchers to quickly determine the state of the cooling system. Measurements of the temperature and overlap of the molecular and atomic clouds make it possible to make judgements regarding cooling efficiency and adjust the system accordingly. Mechanical beam shutters with sub-microsecond rise times can block and unblock beams, speeding up the diagnostic process.

1 Introduction

The production of large amounts of ultracold molecules has potential benefits in many active areas of scientific research. These molecules could allow us to explore quantum phenomena that are not currently understood, as well as help build technologies that have never been possible before. For example, these molecules have the potential to be used as qubits in a scalable quantum computer [1]. Unlike bits, which can represent either the number zero or the number 1, qubits can either represent one of these numbers or be in a superposition state of both of them. This allows a small number of qubits to contain information about many numerical possibilities simultaneously, thus speeding some computations up exponentially. In order for qubits to be useful, they must be have a certain set of properties, including the ability for their quantum states to be easily manipulated. This requirement makes molecules attractive candidates because of their rotational energy states. The energy differences between these states are such that a photon emitted or absorbed in a transition would be in the microwave wavelength spectrum. Because microwave radiation can be easily manipulated by electronic circuits, the same circuits could also control energy state transitions in molecules. It is also necessary that it be possible to initialize the qubits in a common quantum state. This is possible with ultracold molecules because at the lowest possible temperatures they will all be in the quantum ground state.

There are several groups developing techniques to cool molecules to their lowest energy states[2]. Some groups have been successful adapting atomic laser cooling techniques for use with molecules. Others have created ultracold molecules through the photo association of laser-cooled atoms. However, this is not a very general tecnique beacuse only a few types of atoms can be laser-cooled. Laser cooling can only be done on molecules with simple energy structures, and most molecules cannot be easily produced from laser-cooled atoms. Other groups have used atomic ions to cool molecular ions through collisions. This method can be applied to a large variety of molecules, but the coulomb interaction between the atomic and molecular ions limits the interactions between them to long-range collisions. Through these collisions the translational kinetic energy of the molecules can be lowered, but the vibrational and rotational energies will remain unaffected, leaving the molecules with energy levels too high for applications such as quantum computing.

The Hudson group at University of California Los Angeles is currently developing a new technique that will be able to cool a large variety of molecules to their ground energy states. We are using laser-cooled neutral atoms to sympathetically cool molecular ions. The neutral atoms are able to cool the vibrational states of the molecules because there is no repulsion from the coulomb interaction between them. Molecular ions are convenient because they can be trapped in an oscillating electric field[3]. The atoms will be cooled and trapped in the same location as the molecular cloud using a magneto optical trap (MOT).

2 Cooling Methods

2.1 Laser Cooling of Atoms

Laser cooling is simply conservation of momentum on an atomic scale. When an atom absorbs a photon travelling with a velocity opposite of that of the atom, the atom absorbs the momentum of the photon and slows down. If many atoms in a cloud have slowed then that cloud has, by the definition of temperature, cooled down. Because of the quantized energy levels in atoms, the photon will only be absorbed if it is on resonance with an available energy transition in the atom. This means that the laser frequency used in the cooling process must be chosen such that it matches an energy transition in the atoms that are being cooled.

The momentum of each photon is significantly lower than that of each atom, so this process must repeat itself many times for the atoms to reach our desired temperature of less than a tenth of a Kelvin. The requirement of repetition limits the types of atoms and molecules that can be laser cooled. The atom must have a cycling transition with a small time constant. If an electron is excited to the upper level of this type of energy structure, it will quickly decay down to the original energy state where it can absorb another photon of the same wavelength. This is important because it allows a beam of identical photons to excite a single atom repeatedly in a short amount of time, causing significant slowing and therefore cooling.

If an atom moving in the same direction as a photon were to absorb that photon, the atom would speed up and heat the cloud. Thus the cooling mechanism must be designed in such a way that photons are only absorbed by atoms moving toward the laser source. This can be done by taking advantage of Doppler shifting of the light. The photons emitted by the source will have a set energy and frequency as observed in the laboratory frame. Atoms moving towards the source will see photons higher than this energy, and atoms moving away will see photons lower than this energy. Atoms can only absorb photons within a narrow range of frequencies. By setting the laser photon energy slightly lower than is needed for absorption, we ensure that only atoms moving towards the source will observe photons with high enough energy for absorption. With this technique implemented, only atoms moving towards the source will absorb photons, so the lasers will always slow the atoms. By using six orthogonal lasers we can slow atoms moving in every direction in the cloud, creating an optical molasses in which the faster the atoms move, the more quickly they are cooled.

The system of six lasers with energy slightly below the cycling transition can be used to trap the atoms as well as cool them. We do this using a magneto optical trap (MOT) [4]. When the atoms are placed in a magnetic field, this field alters the energy structure of the atom. Levels that were degenerate in the absence of a field now spit into multiple states that are each either higher, lower, or equal to the original energy level. The stronger the field is, the larger this shift becomes. By putting one coil of wire above the desired cloud center and another coil below, then putting current through the two coils in opposite directions, we create two magnetic dipoles. The fields of these dipoles will cancel to zero at the point exactly between them if the currents are of equal magnitude. The strength of the field increases as distance from the center point increases. As a result, the atoms in this field experience a higher energy shift the farther they get from the center. This shift decreases the energy difference between the ground state and the lowest possible excited state. Therefore, the farther an atom is from the center, the more strongly coupled this energy transition will be to the energy of the photons in the lasers. Because of this, all of the atoms will be pushed towards the center of the trap.

In our experiments we are cooling neutral calcium atoms. In these atoms, the highest-energy electrons are in the 4S state. These electrons can be excited to the 4P state with 422 nm light. This transition has a lifetime of about 4.5 ns. However, electrons in the 4P state do not always decay back to 4S. Sometime they decay down to 3D, at which point they cannot decay down to 4S [5]. In order to prevent electrons from getting stuck in a state where they cannot absorb cooling photons, we have a second set of lasers. These lasers emit 672 nm light. Electrons in the 3D state can absorb this light and be excited up to the 4P state, where they can decay back to ground and continue the cooling cycle. The atoms in our system absorb on the order of ten-million photons per second, making it possible to cool the atom cloud to temperatures below a tenth of a Kelvin in only a fraction of a second.

2.2 Sympathetic Cooling of Molecules

The necessity of the cycling transition in laser cooling makes it very difficult to laser-cool most molecules. The energy structure of a molecule is typically much more complicated than that of an atom. Atoms have only electronic energy states. Molecules have electronic states as well as rotational and vibrational states. These additional transitions are in the microwave region, and they are what make molecules potentially useful in quantum computing. However, they are extremely long-lived and non-cycling. Electrons in atoms that quickly decay to the ground state can be consistently excited by a cooling laser. Once excited, electrons in molecules will eventually decay to the ground state but they will most likely decay to other states first and stay for long periods of time in these upper levels. In this situation, the energy transitions available to the electron will be off-resonance with the cooling lasers so the molecules will not absorb photons and cool down.

Sympathetic cooling is a useful molecular cooling method because it does not depend on the energy structure of the molecules. We simply trap a cloud of molecules with a cloud of atoms cooled by well-established laser-cooling techniques (as described above). The molecules and atoms collide because of their close proximity, and in these collisions some of the energy in the hot molecules is transferred to the cold atoms. The fast-moving heated atoms are then kicked out of the MOT. Theoretically, this process should cool the molecules to temperatures close to the laser-cooled atoms. The atoms are neutral, so there is a coulomb attraction between the molecules and the atoms. This allows for short-range collisions that should be able to lower the vibrational and rotational energy of the molecules.

In this method we use molecular ions because they can be trapped in a time-varying electric field. We create this field by placing four conducting rods parallel to each other around the center of the MOT. We then ground two rods that are diagonal from each other in the rectangle, and place an alternating voltage across the other two rods. This creates a saddle-shaped potential which rotates as the voltage alternates from high to low, creating a minimum potential in the center of the four rods. Since the center of the rod setup is aligned with center of the MOT, the molecular ion cloud is trapped in the same location as the neutral atom cloud, and the atoms can sympathetically cool the molecules.

This trapping mechanism results in a large molecular cloud that is elongated in the direction parallel to the rods. Because the MOT cloud is spherical and smaller than the molecular ion cloud, the overlap between the atomic and molecular clouds is too low to significantly reduce the translational energy of the molecules. In order to account for this, we dissociate some of the BaCl+, splitting molecules into Ba+ and Cl. We then laser-cool the Ba+ ions, which are trapped with the BaCl+ molecules in the electric field and thus have better overlap with the molecules. The Ba+ atoms sympathetically cool the translational modes, but cannot cool the inner energy levels of the molecules because of the coulomb repulsion between the molecular and atomic ions. These internal energies are cooled by the neutral Ca atoms. In order to effectively cool the molecules to the absolute ground state, ultracold Ba+ ions and Ca atoms must both be present. One we increase the overlap between the Ca and BaCl+, the Ba+ ions will no longer be necessary.

3 System Diagnostics

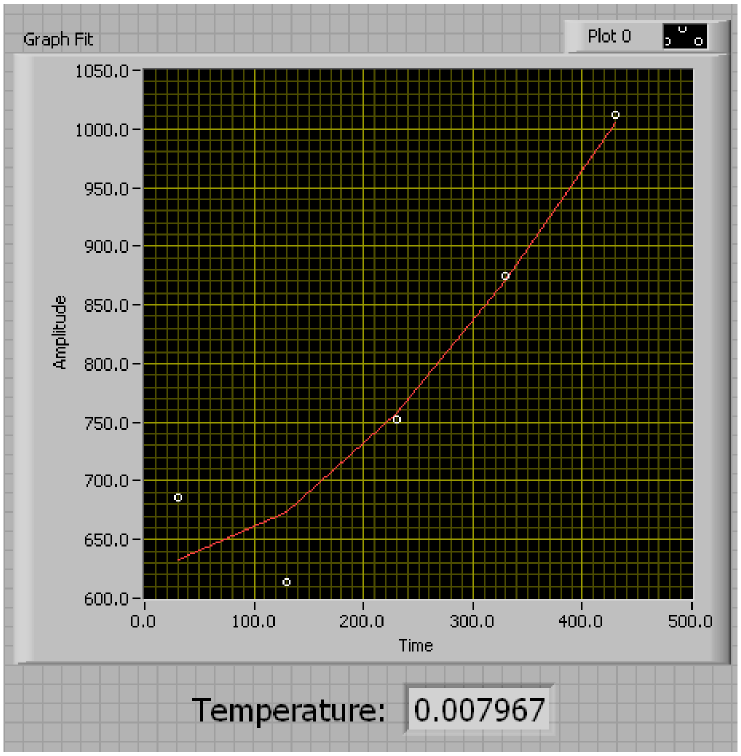
As we continue to improve our trap design and methods, it is important to be able to take quick measurements of the system. Knowing the state of the system allows us to determine the effects of any alterations, and can tell us whether the setup is working properly. Labview programs built to control detectors and laser-related devices make taking these measurements easy and efficient. Also, updating laser-control technologies can improve the speed at which measurements can be taken.

3.1 Temperature

Measuring the temperature of atomic cloud is a good way to determine how well the MOT is working. We can find this information by observing how quickly the cloud expands when we block the cooling/trapping lasers. The size of a gas as it expands into a vacuum depends on three factors: the mass of the particles, the size of the cloud before expansion, and the temperature. We know the mass of Ca, so by turning off the lasers and measuring the cloud at different points in time as it expands, we can make an estimate of the initial cloud size and the temperature. The waist size w(t) is related to the initial waist w_o, the Boltzmann constant k_B, temperature T, mass m and time t in the following way:

$$w(t) = \sqrt{{w_o}^2 + \frac{2k_B T t^2}{m}} \tag{1}$$

We take pictures of the cloud using absorption imaging. This involves turning off the MOT lasers, and turning on an additional absorption laser that is exactly on resonance with the cycling transition in the atom. This laser is oriented so that it passes through the atomic cloud and then into a camera. The width of the laser beam is larger than the size of the cloud. When the photons from the beam pass through the cloud, the atoms absorb many of the photons and leave a shadow on the detector. The darkness of the shadow on the two-dimensional image is proportional to how many atoms the beam passed through to reach that point. We assign values to each pixel such that the darker the pixel is the higher its value, and select a rectangular area of interest that contains the shadow and eliminates noise around the edges of the image. We then use the values within the selected area as weights to calculate the weighted numerical standard deviation of the image. This standard deviation is the waist of the cloud at the time the image was taken.



Temperature: 0.007967

Figure 1: Graph of waist sizes at different points in time, with line fitted according to formula (1), and temperature outputted at bottom.

Once the cloud is no longer trapped by the MOT, it takes about 1 ms to completely fill the vacuum chamber. We need to take several images of the expanding cloud in order to accurately estimate the temperature. Because of the speed of expansion, we choose to analyze five images taken ten microseconds apart. However, our detectors cannot record images this quickly. Instead of taking five pictures within 50 microseconds of each other, we take one picture ten microseconds after the start of expansion and then turn on and reload the MOT. We then turn the MOT off and take the next image after twenty microseconds. We repeat this process for each image. Once we have a set of standard deviations of the cloud at different times, we use Eqn (1) to fit a curve to these points. The best fit gives us the temperature of the atomic cloud. Typically these temperatures range from around six to ten millikelvin.

The temperature of the molecular cloud cannot be determined by absorption imaging because there is no frequency of light that will be absorbed by the molecules consistently enough to cast an observable shadow. We are working on

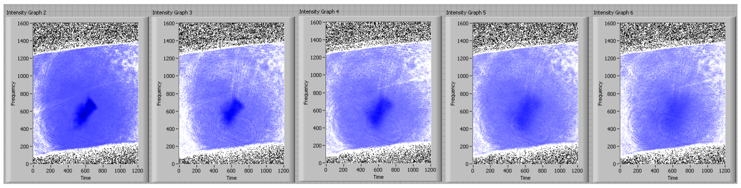


Figure 2: Absorption images of expanding MOT cloud, taken 10 microseconds apart

several different ways of determining molecular temperatures. We can calculate the cooling rate of the molecules due to sympathetic cooling with atoms. However, there is also a heating factor due to collisions between molecules as they move in the oscillating electric field. We are currently building simulations to determine this heating rate. The combination of the heating and cooling rates will be able to tell us some information about the temperature of the atoms. In order to establish whether or not the molecules are in their absolute ground energetic states, which is our ultimate goal, we need to first develop a thorough model of the energy structure of the Barium Chloride ions. We are working on this using spectroscopy.

3.2 Overlap

The effectiveness of sympathetic cooling depends on the occurrence of frequent collisions between the molecules and the atoms. To maximize this collision rate, we need to make the density and overlap between the two clouds as high as possible. The overlap factor ϕ is the fraction of the peak MOT density that the average molecule sees, as illustrated in the Eqn (2).

$$\rho = \phi * \rho_{MOT(peak)} \tag{2}$$

In order to calculate this overlap factor we need to know the size, shape, orientation, and location of each of the clouds. We know the approximate shape and orientation of the clouds because we know how they respond to our trapping mechanisms. In order to determine the sizes and relative positions of the clouds, we take images of the clouds from two different angles. We find the numerical horizontal and vertical standard deviations of these clouds using a method identical to that used in making temperature measurements of atoms. We can also use the two images to triangulate the center of each cloud.

Absorption imaging is not a viable tool for taking the pictures needed to determine the overlap factor. We need to know where the clouds are located while the cooling lasers are on, but leaving them on would add enough background light so that it would be impossible to analyze an absorption shadow. Instead, we use fluorescence imaging. Every time an atom absorbs a photon from one of the cooling lasers, it quickly emits this photon in a random direction. We can detect these photons and use them to create images of the atom cloud.

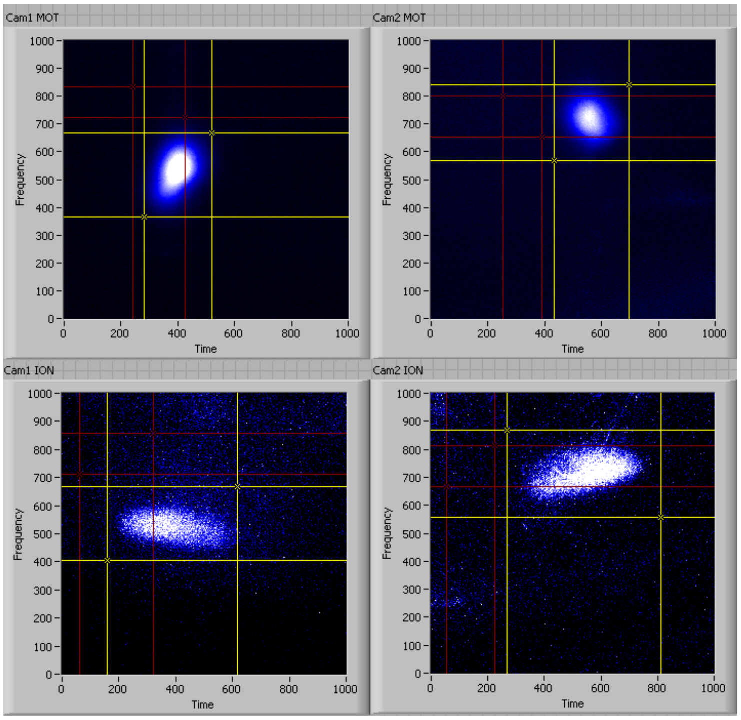


Figure 3: Flourescence images taken for overlap calculation. Top: MOT images from camera 1 and camera 2. Bottom: ion cloud images from camera 1 and camera 2

The molecular ions are not absorbing or emitting photons, so we cannot use fluorescence imaging to see them. In order to determine where they are in relation to the atom cloud, we remove the molecular ions from the trap and replace them with Yb+ atomic ions. These atomic ions have the same mass and charge as the molecular ions, so the electric field trap affects them in the exact same way. Therefore, the atomic ions will be in the exact same place as the molecular ions were. We laser-cool the Ytterbium ions while they are in the ion trap. The atomic ions fluoresce because they are absorbing and emitting light from the cooling laser. We can detect this fluorescence and create an image of what the molecular ions would look like if we could see them in the trap.

Once we have information about the two clouds, we approximate the MOT cloud as a spherical Gaussian and the ion cloud as an ovoid Gaussian. This allows us to create a function representing the density of each cloud. In Eqn (3), w_x, w_y, and w_z are the standard deviations of the cloud in three different directions. For the MOT these three values are the same. The y-axis is chosen so that it is aligned with the elongated direction of the ion cloud. The x and z standard deviations of the ion cloud are equal. The values xo, yo, and zo represent the location of the center of the cloud. The center of the coordinate system is arbitrary but consistent between the two clouds because we are interested only in their relative positions.

$$\rho(x,y,z) \propto exp(-(\frac{x-x_o}{w_x})^2 - (\frac{y-y_o}{w_y})^2 - (\frac{z-z_o}{w_z})^2) \tag{3}$$

The MOT density function is peak normalized and the ion function integral normalized. This means that we will only have an overlap factor of one if the ion cloud is concentrated at the location of the peak density of the MOT. The resulting functions are multiplied together and integrated over all space to find the overlap factor.

$$\phi = \int\int\int d^3\mathbf{r}\rho_{MOT}(\mathbf{r})\rho_{ION}(\mathbf{r}) \tag{4}$$

Typical overlap factors for our current design are between ten and twenty percent.

3.3 Shutter

In the process of developing and testing this cooling method, there are many beams of photons, atoms and molecules that are being manipulated. In many cases, the turning on and off of beams must be done on microsecond times scales. This makes it important that we have the fastest, most efficient methods of manipulating these beams.

Scientists at the University of Melbourne have designed a shutter that can mechanically block and unblock beams with nanosecond consistency and rise times of less than a microsecond [6]. This shutter also costs almost no money to build, as it can be made out of the magnet and voice coil in a broken hard drive. In the hard drive, the voice coil is placed on top of a bar magnet. When a current is sent through the coil there is a force on the coil due to the interaction between the current and the magnetic field. If this current is reversed, the force switches to the opposite direction. We can create a simple circuit using an H-bridge that can quickly switch the direction of current in response to an electronic signal. If we connect a blocking mechanism to the voice coil, it can be quickly moved back in forth to block and unblock a beam.

When we built this device, we found that if the beam is small enough, we can achieve rise times on the order of a microsecond. The electronic signals are have a reliability on the order of a microsecond, meaning that if a pulse is sent to the circuit, we can predict when the circuit will output a signal to the voice coil with microsecond accuracy. However, the mechanical shutter itself is unreliable. In the current shutter design, the beam passes through a small slit as the shutter moves up or down, creating millisecond pulses. The length of these pulses can be controlled by focusing the beam so it passes through different parts of the slit. The shutter can also block the beam for millisecond length scales if the beam above the arm of the shutter. As the shutter moves up and down the arm passes over the beam, blocking it for a few milliseconds. This design allows for significant flexibility in the use of the shutter, but the shape

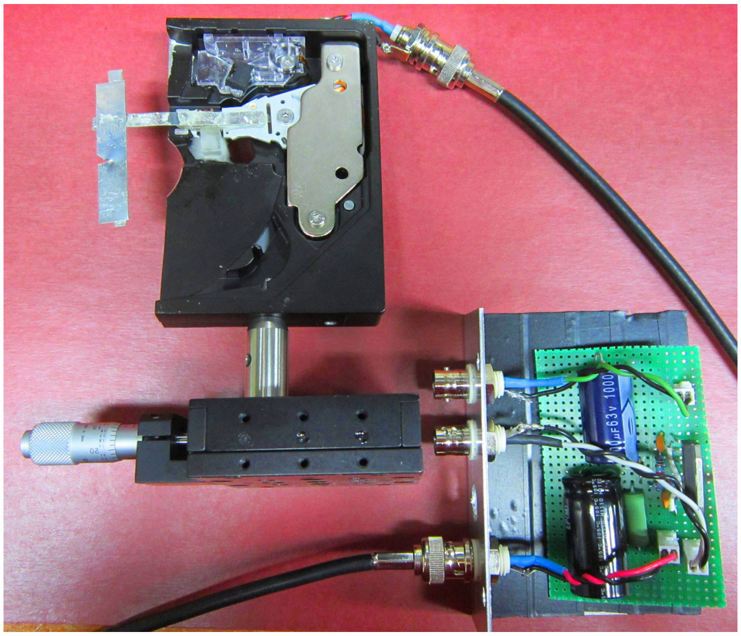


Figure 4: Shutter built from voice coil of broken hard drive
The blocking mechanism is on the upper left, connected to the arm that moves up and down in response to changes in the voice-coil current. On the right is the h-bridge circuit that controls the current through the voice coil.

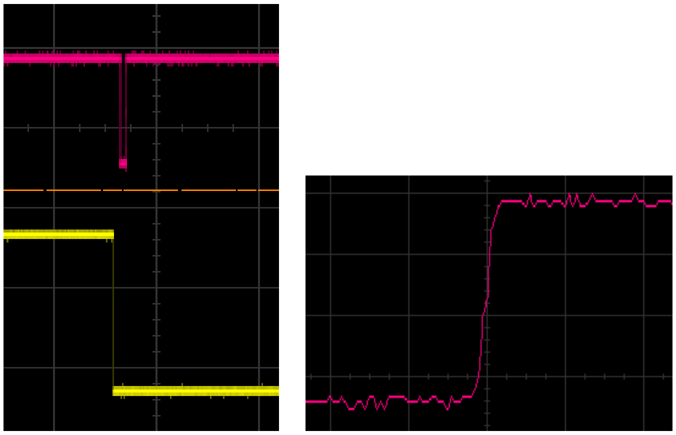
and material result in significant vibrations that damage the arm and make it unreliable. The arm often gets stuck on both its upswing and downswing for reasons that are not apparent. The components of the shutter are held together with epoxy, which is not strong enough to withstand the forces on the arm as it is stopped by bumpers.

In order for this new technology to be useful in our lab, a new shutter arm would have to be designed. The pieces would have to be held together in a more permanent way than epoxy. A set of screws may be effective. The blocking mechanism would have to be designed in such a way that vibrations are damped. This could mean using a different, less flexible material. Also, it would be important to find out why the arm gets stuck, and to correct for this problem.

4 Conclusions

The sympathetic cooling of molecular ions using laser-cooled atoms is a promising new molecular cooling method. The future of this research lies in improving the trap design so as to increase the overlap between the atoms and molecules. Further research regarding the heating rate and energy structure of the BaCl+

Figure 5:



(a) Shutter moves in response to signal, blocking the laser for a brief interval as the arm passes between the beam and the detector. Top signal: output from photodetector. Bottom signal: voice coil input

(b) The rise time is the time interval between when the shutter begins unblocking the beam and when the beam is completely unblocked. This value is determined by the speed of the shutter and the width of the beam.

molecular ions we are working with will allow us to determine how well this method works.

The new Labview tools for measuring temperature and overlap of the cooling system are efficient, easy to use, and accurate. The mechanical shutter is a potentially useful tool, but would require more testing and a redesign of the blocking mechanism.

5 Acknowledgements

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