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Micro-Kelvin Cold Molecules

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Abstract

We have developed a novel experimental technique for direct production of cold molecules using a combination of techniques from atomic optical and molecular physics and physical chemistry. The ability to produce samples of cold molecules has application in a broad spectrum of technical fields high-resolution spectroscopy, remote sensing, quantum computing, materials simulation, and understanding fundamental chemical dynamics. Researchers around the world are currently exploring many techniques for producing samples of cold molecules, but to-date these attempts have offered only limited success achieving milli-Kelvin temperatures with low densities. This Laboratory Directed Research and Development project is to develops a new experimental technique for producing micro-Kelvin temperature molecules via collisions with laser cooled samples of trapped atoms. The technique relies on near mass degenerate collisions between the molecule of interest and a laser cooled (micro-Kelvin) atom. A subset of collisions will transfer all (nearly all) of the kinetic energy from the "hot" molecule, cooling the molecule at the expense of heating the atom. Further collisions with the remaining laser cooled atoms will thermally equilibrate the molecules to the micro-Kelvin temperature of the laser-cooled atoms.

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Summary

The Micro-Kelvin Cold Molecule Laboratory Directed Research and Development project's primary objective was to develops a new experimental technique for producing micro-Kelvin temperature molecules via collisions with laser cooled samples of trapped atoms. Recently, experiments with cold atoms have demonstrated surprising versatility as the primary technology behind a new generation of technical devices (clocks, gyros, magnetometers, quantum gates) and new insight into fundamental science (dynamics, quantum statistics, quantum electro-dynamics, super-fluids high temperature superconductors). Performing similar research with cold molecules promises a much broader spectrum of applications due to the complex structure of molecules. This complex structure of molecules greatly enhances the potential of these systems, but these complexities makes it impossible to use the same cooling techniques available for making cold atom samples (laser cooling). There are several techniques that are being pursued around the word for producing samples of cold molecules, however, they rely on specifi internal characteristics of a small class of molecules at a construction cost of several million dollars.

Here at Sandia National Laboratories, using ideas based on past experiments funded by the Department of Energy, Offic of Basic Sciences, we have developed a new, broadly applicable, low-cost, technique for producing samples of cold molecules. Single collisions between near mass degenerate particles will exchange kinetic energy between the particles leaving one with no kinetic energy and the other with all the kinetic energy. The particle with no kinetic energy is stationary (no velocity) in the laboratory and hence cold, as temperature is proportional to the velocity squared. This kinetic (collisional) cooling technique is extremely general; it works for atoms and molecules and is independent of all internal properties except the mass of the particle. Using this simple but clever technique in combination with laser cooling of atoms to micro-Kelvin temperatures we have developed and demonstrated the basic technique for cooling molecules from supersonic velocities ($\sim$ 900K) to a millionth of a degree above absolute zero. These cold samples of molecules provide for a fundamentally new approach in chemical dynamics, in that we can now control and manipulate chemical systems on the quantum level.

Nomenclature

MOT Magneto-Optical Trap

Trap any device that spatially localizes atoms or molecules without walls

EO Electro-optical modulator: a device that frequency modulates a laser beam allowing multiple narrow transitions to be driven with a single beam.

AO Acousto-optical modulator: a radio-frequency device that shifts the optical frequency of a laser beam.

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1 Inroduction

The cooling of molecules to sub-Kelvin temperatures promises to have a great impact on the field of chemistry and physics. At these temperatures, the molecules are moving much more slowly in the laboratory reference frame compared to typical molecular beam velocities, leading to significantl longer observation times. At these low kinetic energies, external electric, magnetic, or optical field can be used to trap, store, and manipulate the molecules, making them ideal targets for studying collisional dynamics, femto/atto-second time resolved spectroscopy, and radical-radical (barrierless) reactions. The ability to confin and control the molecules along with the extended observation times allows for ultra-high resolution spectroscopy that, for example, may enable the direct counting of the density of states of the molecules[2], as well as precision measurements of fundamental physical constants [13, 26, 11], and even the detection of gravitational waves [32]. By trapping and orienting molecules prior to photo-dissociation, the spatial averaging associated with randomly oriented molecules can be greatly reduced, yielding new details in the observed angular distributions of the photo-fragments that can reveal the underlying dynamics of the photo-dissociation process [30]. Another consequence of the slow molecular velocities at these low temperatures is the increase in the de Broglie wavelength associated with the molecules. In analogy to atom optics, the wave-like properties of the cold molecules can be exploited to perform unique interferometric studies with molecules, such as the separation of mixtures of clusters by diffraction through a transmission grating [25, 14, 3].

To date there has been limited success in producing samples of cold molecules. Two general strategies are being used: welding together pre-cooled atoms into diatomic molecules, and direct slowing or cooling of a molecular beam. The predominant techniques used to create cold molecules from cold atoms are photo-association [33, 23] and Feshbach resonance association [24, 29]. Photo-association of atoms consists of optical excitation of the unbound atoms to a bound, long-range electronically-excited molecular state, followed by either Frank-Condon emission from that excited state to bound levels of the ground electronic state, or laser stimulation from the excited state to a particular ground vibrational state. In Feshbach resonance association, magnetic field are used to tune the energy of a colliding pair of atoms in coincidence with a bound singlet state of the diatomic molecule. The primary advantage to welding together pre-cooled atoms is that the molecules formed are born translationally cold, typically in the micro-Kelvin temperature range. However, the molecules are formed in highly vibrationally excited states that can rapidly quench via collisions. Recent experimental advances[6, 22, 31] have utilized optical pumping schemes to transfer molecules to more deeply bound states, including the absolute ground state. These techniques apply only to diatomic molecules that are comprised of a small selection of atoms that can be laser cooled, primarily alkali metals.

Several techniques have been suggested for generating cold molecules by direct slowing or cooling of faster moving molecules. In these experiments, the molecules are typically injected into a high-vacuum region using a molecular beam valve. The supersonic expansion of the molecules into the vacuum region cools them down to their ground vibrational state with a rotational distribution of a few degrees Kelvin. While the internal degrees of freedom are cooled by the expansion, the molecules are left translationally hot, moving at supersonic velocities in one dimension. The

goal of the direct cooling/slowing is to remove this translational energy and bring the molecules to rest in the laboratory reference frame. Direct slowing of molecules has the advantage that a larger variety of diatomic and complex molecules may be cooled.

One conceptually straightforward technique for slowing down molecules in a supersonic beam is to move the molecular beam valve backwards in the laboratory reference frame with a velocity equal and opposite to that of the escaping molecular beam, canceling out the translational velocity in the laboratory frame [10]. Other techniques fall broadly into three categories: using external electric or magnetic fields using optical fields or using collisions. Both Stark deceleration [1, 18] and Zeeman deceleration [12, 21] have demonstrated the successful production of samples of cold atoms and molecules using external electric and magnetic fields respectively. Optical fiel techniques include the use of far off-resonance laser beams for optical Stark deceleration [9] and the production of very polarizable molecular Rydberg states [27] that can be easily slowed with electric fields Collisional techniques capable of producing molecules at sub-Kelvin temperatures include bulk cooling using 300 milli-Kelvin helium gas as a collision partner [20], the entire fiel of trapping molecules in superfluidi helium clusters[4, 5], and kinematic cooling. In kinematic cooling, molecules undergo single collisions with atoms such that a subset of the molecules are left moving slowly in the laboratory reference frame [8]. It is so called because the amount of cooling is strictly determined by the kinematics of the collision. The possibility of creating a cooled molecule as the product of a reactive scattering event has also been investigated[19]. These techniques have produced samples of molecules in the milli-Kelvin temperature range at densities that are too low to be considered ultracold.

Here we present a novel technique based on kinematic cooling physics, which offers greater experimental simplicity than the current kinematic cooling technique. In conventional kinematic cooling, supersonic molecules in a molecular beam are cooled to milli-Kelvin temperatures by either elastic [16] or inelastic [28] collisions with an atomic beam intersecting at a near 90 degree angle. Recent experiments[28] have shown that secondary collisions between the cooled atoms and the remnants of the parent atomic and molecular beams can reheat and destroy the cold molecule sample. Here we propose replacing the target supersonic atomic beam with a dense cloud of pre-cooled atoms in a magneto-optical trap (MOT). This seemingly minor change alters the kinematic equations governing the cooling process, affecting the experimental geometry and allowing for a surprisingly large number of molecules to be cooled to sub-Kelvin temperatures via elastic scattering. Further, secondary elastic collisions with the atoms would no longer collisionally reheat the cold molecular sample, but rather serve to further cool the molecules.

2 Backround

The physics behind this novel cooling approach can be summed up in the behavior of a Newton's cradle device. Consider a Newton's cradle with two balls of equal mass; one ball is initially stationary while the second gets pulled and released. As the second ball strikes the firs head on, all of the kinetic energy from the second ball is transferred to the first bringing the second ball to rest. This is analogous to the kinematic cooling process, wherein a "hot" molecule collides with a near stationary atom of similar mass, ejecting the atom and bringing the molecule to rest. The difference is that in the Newton's cradle, the balls have equal mass and are constrained to collide with a zero impact parameter. To generalize this to molecules colliding with laser cooled atoms, we must analyze the collision between these particles for arbitrary masses and non-zero impact parameters.

The collisional process can be characterized by the amount of energy transferred from one particle to another, which we calculate by modeling the colliding particles as hard spheres. Consider two particles: particle A with mass m_A and velocity v_A, and particle B with mass m_B and velocity v_B. Figure 1 shows the relationship between the velocity v_A of particle A in the laboratory frame of reference and the velocity u_A in the center-of-mass reference frame. Primed and unprimed vectors refer to post- and pre-collision velocities, respectively. After the collision, the particle scatters onto a sphere in velocity space (dictated by conservation of energy and momentum) around the center-of-mass (COM) of the system, which moves with velocity $\overrightarrow{v}_{COM} \equiv [m_A \overrightarrow{v}_A + m_B \overrightarrow{v}_B]/(m_A + m_B)$ in the laboratory reference frame. The scattering angle χ is determined by the impact parameter of the collision.

For elastic scattering,$U_A = U'_A$, and the product velocity vectors have the following relationship,

$$(v'_A)^2 = (U'_a)^2 + v_{COM}^2 - 2\cos(\pi - \chi) \cdot v_{COM} \cdot U'_A \tag{1}$$

Assuming particle B is initially at rest (v_B=0), the scattering angle in the center-of-mass frame is given by

$$\cos(\chi) = [(v'_A/v_A)^2 - (m_A^2 + m_B^2)/(m_A + m_B)^2]/[2\mu_{AB}/(m_A + m_B)], \tag{2}$$

where μ_{AB} is the reduced mass of the system. For equal masses, this reduces to $\cos(\chi) = 2(v'_A/v_A)^2 - 1$. We see that the post-collision velocity of particle A in the laboratory frame is minimized for large scattering angles.

Figure 2 shows the relationship between the scattering angle and impact parameter of the collision

$$b = IB = d\cos(\alpha) = d\cos(\chi/2), \tag{3}$$

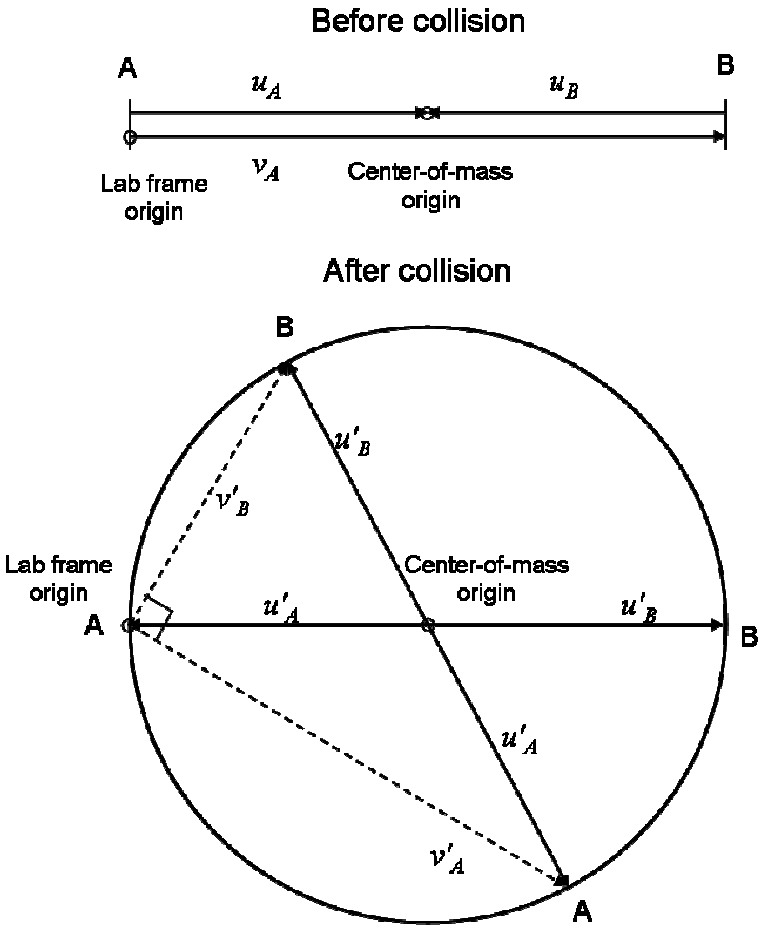

Figure 1. Vector diagrams of the pre- and post-collision velocities for a hard sphere collision between two particles with equal masses. Initially, particle *A* has velocity v_A and particle *B* is at rest in the laboratory frame (lab frame velocities are designated by *v*). In the center-of-mass frame, half of the velocity is in *A* and half is in *B* (center-of-mass velocities are designated by *u*). After the elastic collision, conservation of energy and momentum require that the fina velocities of *A* and *B* (designated as primed) lie on a sphere in velocity space as shown above where $|u_A| = |u_B| = |u'_A| = |u'_B|$. The intersection between the sphere and lab frame origin corresponds to the case where $v'_A = 0$. In the laboratory frame the particles *A* and *B* are scattered at 90°.

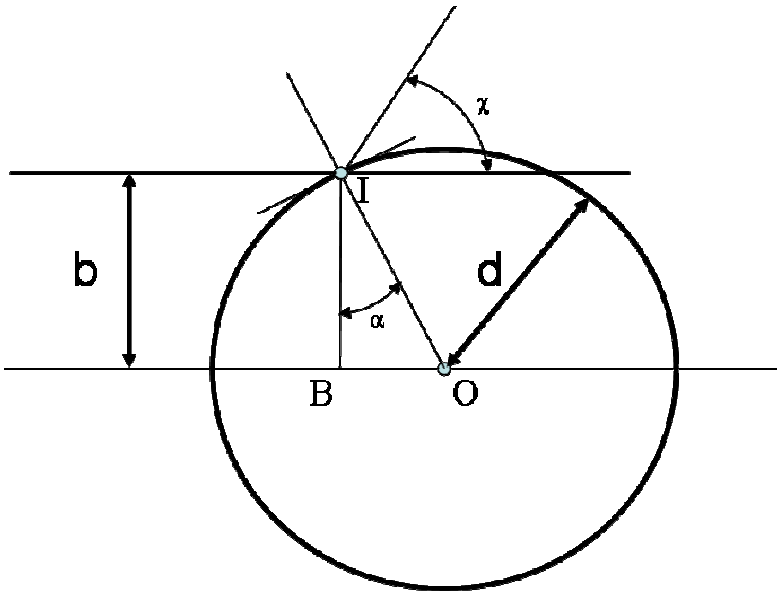

Figure 2. Dynamics of two-particle hard sphere scattering. The center-of-mass frame scattering angle is given by χ, and α is the angle subtended by the radius to the impact point and the perpendicular to the center line of the collision. The impact parameter is given by b, and d is the sum of the radii of the two spheres.

where $d = R_A + R_B$ is the distance of minimum approach between the center-of-masses of the two spheres. From this one-to-one correspondence between the angle of scattering and the impact parameter we see that the post-collision velocity of particle A is minimized for small impact parameters. This is expected, as for a head-on collision b = 0 and $\chi=\pi$ resulting in direct recoil, while for $b=d$ and $\chi = 0$ no deflectio is expected.

To determine the number of cold molecules that can be generated using this kinematic cooling technique, we firs need to fin the partial differential cross-section $\sigma_{p(min)}$ for collisions resulting in a minimum scattering angle χ_{min}, from which we can calculate the fraction of collisions, $F \equiv \sigma_{p(min)}/\sigma_T$, that cool the molecules down to the corresponding maximum fina velocity. The partial differential cross-section is determined by summing contributions from impact parameters between zero and b_{min}

$$\sigma_{p(min)} = \int_0^{2\pi}\int_0^{b_{min}} bdbd\phi = \pi d^2\cos^2(\frac{\chi_{min}}{2}) = \pi d^2(\cos(\chi_{min}+1)/2), \tag{4}$$

while the total cross-section of all collisions is given by the total area of the collision,

$$\sigma_T = \int_0^{2\pi}\int_0^{d} bdbd\phi = \pi d^2. \tag{5}$$

The fraction of collisions having $\chi \geq \chi_{min}$ is therefore $F \equiv \sigma_{p(min)}/\sigma_T = \frac{1}{2}(\cos(\chi_{min})+1)$. Substituting in equation 2 gives

$$F = \frac{1}{2}\{[(v'_A/v_A)^2 - (m_A^2+m_B^2)/(m_A+m_B)^2]/[2\mu_{AB}/(m_A+m_B)]+1\}. \tag{6}$$

Using the fact that $v_A'^2/v_A^2 = E'_A/E_A$, this fraction can be expressed in terms of the initial (E_A) and post-collision (E'_A) energies of particle A,

$$F = \frac{1}{2}\{[E'_A/E_A - (m_A^2+m_B^2)/(m_A+m_B)^2]/[2\mu_{AB}/(m_A+m_B)]+1\}. \tag{7}$$

In the case of degenerate masses $m_A = m_B$, this equation reduces further to

$$F = E'_A/E_A. \tag{8}$$

This simple result gives the fraction of collisions between hard spheres of equal mass that transfers a particular amount of energy, where particle B is assumed to be initially at rest (v_B = 0). Thus, if we require at least half of particle A's initial energy to be transferred to the previously stationary particle B (E'_A = 0.5 E_A), then the fraction of collisions satisfying this condition is also a half. If we want to have a collision that transfers at least 99% of the energy to particle B, then E'_A = $0.01E_A$ and F = 0.01, so 1% of the random hard sphere collisions will accomplish this. For unequal masses, the fraction of contributing collisions decreases by a factor of two when the mismatch of masses exceeds 15%. As a result, this process is capable of slowing a large variety of molecules even for a single target species of atom in the MOT.

The density of cold molecules we expect to achieve with this technique is determined by the density of atoms in the MOT and the fractional energy that needs to be removed from the molecules. At steady state, the rate of production of cold molecules resulting from collisions between thermal molecules and cold atoms is equal to the loss rate due to collisions between the cold molecules and the incoming thermal molecules. We assume that the total cross-section for molecule-atom collisions is the same as for molecule-molecule collisions; however, only a small fraction $F \equiv \sigma_{p(min)}/\sigma_T$ of the molecule-atom collisions results in cold molecules with sufficientl slow velocities in the laboratory frame. Since the rate of collisions is proportional to the density of the particles involved, the density of atoms n_atom and molecules n_mol are related as

$$n_{mol} = F \cdot n_{atom} \tag{9}$$

For example, if the fraction of collisions that sufficientl cool a molecule below a certain temperature is F = 10^{-3}, then a cold molecule will have an equal probability of being generated or lost

when the density of molecules is 10^{-3} that of the atoms. Note that slower initial molecular velocities and higher atom densities in the MOT will increase the number of cold molecules available when steady state is achieved.

This assumes that the main loss mechanism from the trap is collisions with the background molecular gas. If, on the other hand, the density of the background molecular gas is too high, then the equilibrium MOT density n_{atom} will decrease, leading to the formation of fewer cold molecules. In the case where the loss rate of both the cold molecules and the MOT are due exclusively to collisions with the thermal molecules and other background atoms, the equilibrium density of cold molecules becomes

$$n_{mol} = F \cdot n_{MOT} \cdot \frac{\beta}{(1+\beta)^2}, \tag{10}$$

where n_{MOT} is the MOT density in the absence of the additional thermal background of molecules, and $\beta = n_{thermal}/n_{bg}$ is the ratio of thermal molecules ($n_{thermal}$) to other background atoms (n_{bg}). This has a maximum of $n_{mol} = F \cdot n_{MOT}/4$, when $\beta = 1$ or $n_{thermal} = n_{bg}$. In general, however, the loss rate for the MOT is dominated by other sources, such that the thermal molecule density can be increased relative to that of the other background gases without significantl impacting the MOT. In this regime, the density of cold molecules will be much closer to $n_{mol} = F \cdot n_{MOT}$.

This cooling technique has many advantages over other collisional cooling techniques. Unlike buffer gas cooling [7] where the large mass mismatch requires the molecules to undergo multiple collisions to be cooled, this technique relies on single collisions, and therefore does not require having favorable elastic to inelastic scattering cross-section ratios. At typical MOT densities, multiple cooling collisions between disparate mass particles are highly unlikely to occur. Also, in contrast to previous conventional kinematic cooling experiments [28], this technique utilizes elastic collisions and is more efficien for slower initial velocities. Another consequence of using cold atoms as the collisional targets rather than a supersonic atomic beam is that it relaxes the experimental requirement of the 90° scattering geometry, eliminating the need for a well-collimated molecular beam source. This allows for experimental simplification and, as we will discuss below, for much lower temperatures to be achieved.

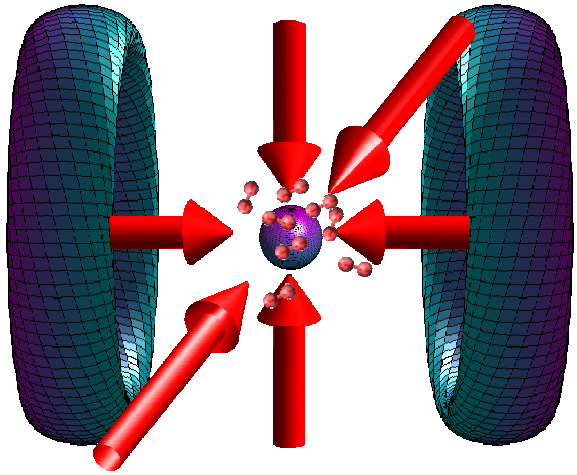

Figure 3. A schematic depiction of the MOT experiment. The red arrows represent the MOT laser beams. The molecules undergo billiard like collisions with the cold atoms, transferring kinetic energy to the atoms, thus slowing the molecules.

3 Experiment

3.1 Methods

The usefulness of any cooling method is largely determined by the total number of trappable molecules that it can generate at a given temperature and density. The micro-Kelvin cold molecule technique, begins with a continuously loaded atomic MOT, into which a molecular gas is introduced with the same or similar mass to the atoms in the trap. The ideal molecular source is one that is both translationally and rotationally cold, and has a high flux A molecular beam provides a source of rotationally and vibrationally cold molecules, but because they are moving at supersonic speeds, the fraction of collisions removing enough kinetic energy to trap the molecules is relatively small. This fraction could be greatly increased by employing more conventional slowing techniques to achieve a lower initial velocity, but at the expense of added complexity. Alternatively, we can consider bleeding the gas of interest into the chamber, which allows for slower initial velocities and consequently colder fina temperatures, as well as a much simpler set up. Because these molecules have a thermal distribution of internal rotational and vibrational states, only a fraction of the cold molecules formed will be in the rotational ground state, and secondary collisions between the rotationally excited molecules and the cold atoms can expel them from the trap. This will reduce the total number of trappable cold molecules at equilibrium; however, the post-collision rotational state distribution itself may provide information about rotational energy transfer and chemical dynamics.

For simplicity, we consider here the case of bleeding in a background gas of the desired mole-

cule. Figure 3 shows a schematic depiction. If the background pressure is low enough not to overwhelm the loading rate of the atomic MOT, then the system will reach equilibrium with some steady state production rate of slow molecules. By superimposing an appropriate molecular trap over the MOT region, we can confin and collect the cold molecules that are generated with energies less than the molecular trap depth. For typical systems, the upper limit of the trappable velocity distribution corresponds to molecular temperatures below 500 mK but greater than 100 mK. Since the cooling efficien y is tied to the relative masses of the colliding particles, we will consider both a mass resonant system and a system with non-equal masses in evaluating the utility of the kinematic cooling technique.

As an example of a mass-resonant system, we consider the scattering of rubidium deuteride ($D^{85}Rb$) molecules off of rubidium 87 (^{87}Rb) atoms collected in a MOT. The cooled $D^{85}Rb$ molecules are to be confine by an optical dipole trap superimposed on the MOT region. The ^{87}Rb MOT is loaded with a typical density of around 10^{10} atoms cm^{-1}, and a background of room temperature (300 K) $D^{85}Rb$ molecules (v_A = 170 m/s) is introduced. If we require a fina velocity of $v_{A'} \sim 14$ m/s (1 K), we fin that $\sim 6.7\ 10^{-3}$ of the collisions produce a $D^{85}Rb$ molecule in this temperature range, giving a steady state density of approximately $6\cdot 10^7$ molecules at sub-Kelvin temperatures. According to equation 6, $\sim 10^{-5}$ of the collisions cool the $D^{85}Rb$ to 10 mK or less, yielding an estimated 10^5 molecules-cm^{-3} in the milli-Kelvin temperature range. Because of the overlap between the MOT and molecular trap, if the collisional losses are favorable, the trapped cold molecules will undergo thermalizing collisions with the ^{87}Rb atoms, which are at a temperature of $\sim 100\ \mu$K. These results are for the most probable velocity in the thermal distribution of the $D^{85}Rb$.

We now turn to a molecule-atom system with a slight mismatch in the particle masses. In the case of deuterated hydrogen bromide ($D^{81}Br$) scattering off of rubidium 85 (^{85}Rb), the initial conditions are similar to that of $D^{85}Rb$ and ^{87}Rb, except that $m_A < m_B$. For the non-mass-resonant system, a single collision cannot bring the molecule to rest in the laboratory frame, and there is a minimum velocity beyond which a molecule cannot be cooled. For example, given a mass ratio of 83 to 85, we fin that $\sim 3\cdot 10^{-3}$ of the collisions can produce $D^{81}Br$ with a fina temperature of 1 K or less; however, no elastic collisions can cool $D^{81}Br$ from an initial velocity of 170 m/s down to temperatures of less than 10 mK. Fortunately, as illustrated in f gure 3a, lowering the initial velocity of the incoming molecules also lowers the minimum obtainable post-collision temperature. If $D^{81}Br$ is cooled to liquid nitrogen temperatures (90m/s), the decrease in the initial velocity shifts the minimum obtainable temperature to a few milli-Kelvin. Figure 3b shows the velocity distribution for a room temperature sample of $D^{81}Br$, and a sample cooled to liquid nitrogen temperatures. Note that in either case, the lower end of the thermal Boltzmann distribution will cool more efficientl to even lower temperatures.

The ability to use an effusive or thermal molecular source lets this cooling technique take advantage of lower impact velocities, allowing it to cool molecules to very low temperatures even with significan mass differences. For experiments where the thermal rotational state distribution of the molecules presents a problem, using this technique in conjunction with other molecular beam cooling techniques such as the counter-rotating nozzle [10] has promise to create samples of cold molecules in an experimentally simple and low cost apparatus. It should be noted that in this

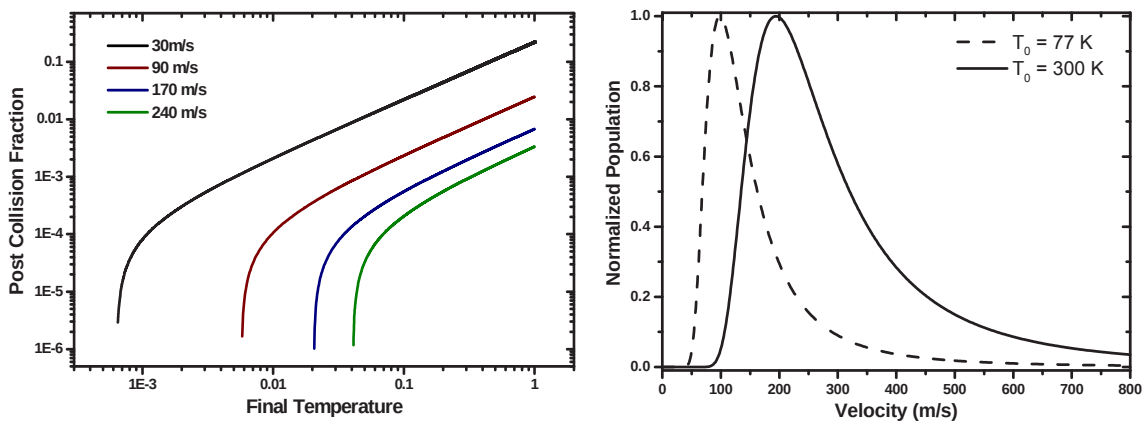

Figure 4. The left hand figur shows the post-collision fraction and fina temperature for selected initial velocities for D81Br scattering off of ^{85}Rb. The right hand figur shows two Boltzmann distributions; the dashed line corresponds to D^{81}Br at liquid Nitrogen temperatures, the solid line is a room temperature distribution, and the vertical line is approximately the temperature of D^{81}Br supersonically expanded in a Xenon carrier gas in a molecular beam.

discussion we have neglected the effects of inelastic collisions with either ground or electronically excited ^{87}Rb on the density of cold molecules that can be formed. These inelastic collisions with electronically excited ^{87}Rb are very exothermic and detrimental to the cooling process but can be greatly reduced by using, for example, a dark-spot MOT [17]. We have also assumed that chemical reactions between the molecules and atoms that can readily eject molecules from the trap are negligible.

3.2 Apparatus

The core technology behind this project is the magneto-optical trap (MOT), which is housed in a ultra-high vacuum chamber. The MOT is formed by three sets of circularly-polarized counter-propagating orthogonal laser beams superimposed over a weak quadrapole magnetic field The trapping lasers are detuned slightly to the red (lower energy) of a ground state atomic cycling transition. The laser detuning and magnetic fiel ensure that the atoms primarily absorb light only from the laser beam the atom is moving towards. This creates a velocity dependant damping force that cools the atoms down to a few cm/s, and spatially localizes them at the zero of the magnetic field Densities in a MOT are typically 10^{10} atoms-cm^{-3} with a spatial size of 1 cm^3. Figure 5 shows a false color image of a trapped cloud of cold atoms and a photograph of the apparatus.

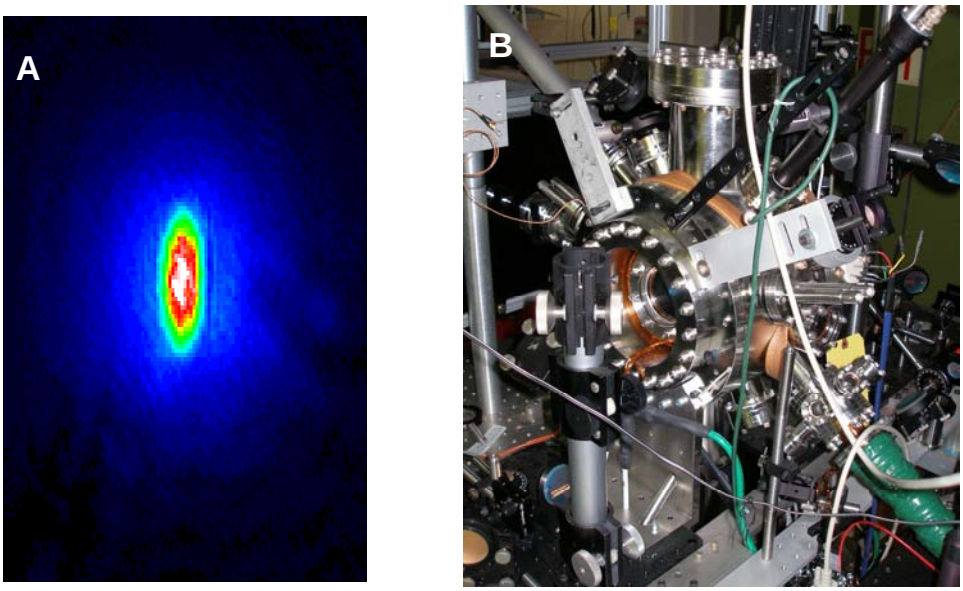

Figure 5. A: A false color florescenc image of a sample of cold atoms in a MOT at a temperature below 0.00001 degree above absolute zero. B: Photograph of the MOT apparatus.

Laser System

The laser system is constructed from two extended cavity Littrow grating diodes lasers from Sacher laser GMB. The master laser outputs 300mW at 780nm, while the secondary laser outputs 120mW at 780nm.

The diode lasers are electronically locked to a Rb vapor cell containing a natural mixture of both isotopes. The lasers are locked directly to hyperfin lines of Rb using a Doppler free saturated absorption setup in combination with a Pound-Drever-Hall technique at 10MHz to remove low frequency noise. The signal is then processed through a homemade PID (proportion integral differential) control circuit. This setup stabilizes the laser frequency to $<$1MZh for up to 18 hours. Longer time may be achievable, but are so far unnecessary.

The rest of the laser light is then modulated to produce both trapping and re-pumping light. Figure 6 shows the four frequencies necessary for trapping both isotopes of Rb. For both of the setups, the re-pumping light is generated using an electro-optical modulator (EO) at 3GHz for ^{85}Rb and 6.8GHz for ^{87}Rb. Acousto-optical modulators (AO) are used to fine-tun the laser frequency and simultaneously act as optical switches, containing the laser light.

The laser light is then coupled to the MOT apparatus by single mode optical fibers The beams are then expanded to a beam waist of 1.4cm, collimated, and broken into six independent beams. Figure 7 show the optical setup for the generation of the six independent laser beams needed to form the MOT. Each beam is then circularly polarized, and steered into the MOT chamber where

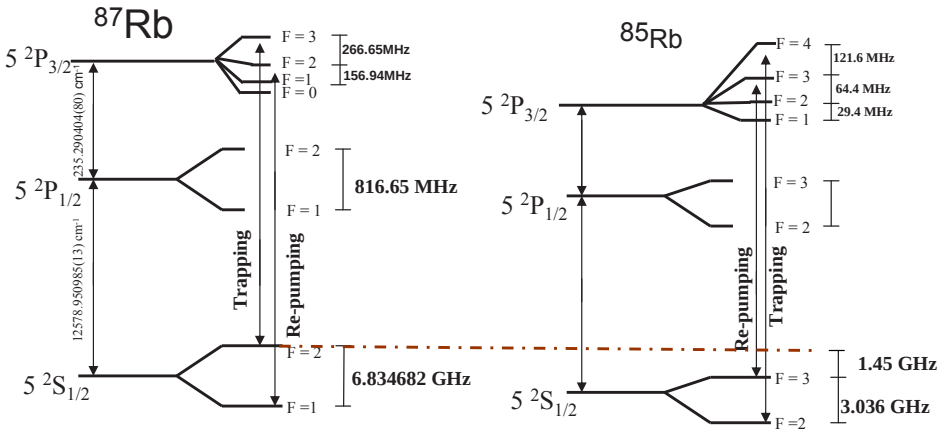

Figure 6. Level diagrams for ^{85}Rb and ^{87}Rb, with the trapping and re-pumping transitions labelled.

they intersect each other in counter-propagating pairs at 90 degrees.

3.3 Imaging

The largest experimental obstacle to to the proof of principle experiment of kinematically cooling ^{85}Rb with ^{87}Rb is distinguishing between the two isotopes upon detection. Figure 6 shows both level diagrams on a relative energy scale. The dashed line that connects the two shows that the isotope shift is on the order of 1.45 GHz. Thus any detection scheme must be able to distinguish energies on this scale.

The primary type of imaging that can be used at the predicted densities for the kinematically cooled ^{85}Rb is florescenc imaging. This causes the main difficult . Atoms in a MOT (the ^{87}Rb are constantly fluorescin due to the trapping and re-pumping lasers. Optical filter need several nm to distinguish between colors, and a monochrometer needs a collimated, well define input. Thus it is challenging to make isotope selective florescenc measurements.

To solve the problem of making isotope selective florescenc measurements we employed a technique of atomic filtering By placing an isotope enriched sample of ^{87}Rb in the imaging system, we can selectively absorb light emitted from resonant ^{87}Rb transitions. The results of the bench test of this system are shown in figur 8.

Using the atomic filte in conjunction with the florescenc imaging allows us to discriminate between signals from the two isotopes. However, this filte does not block far off-resonant excitation signals. This “leakage” signal could be contributing or even obscuring the actual signals.

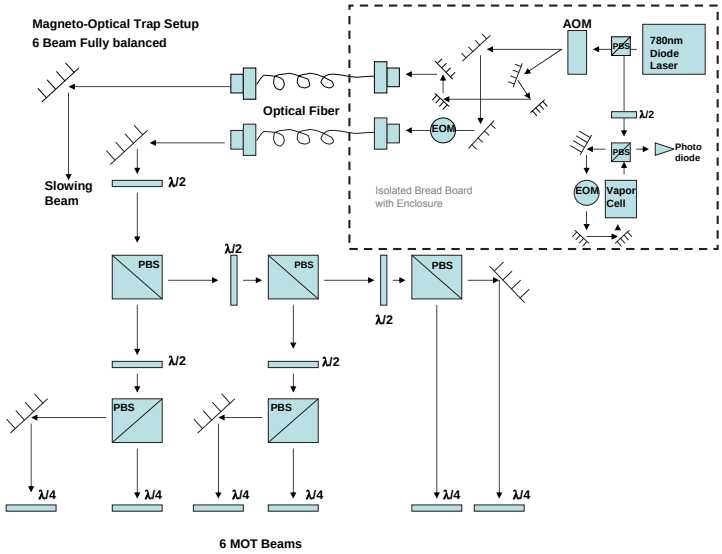

Figure 7. Schematic for the generation of the six laser beams needed for one isotope. In total, four such setups are required for the dual rubidium MOT.

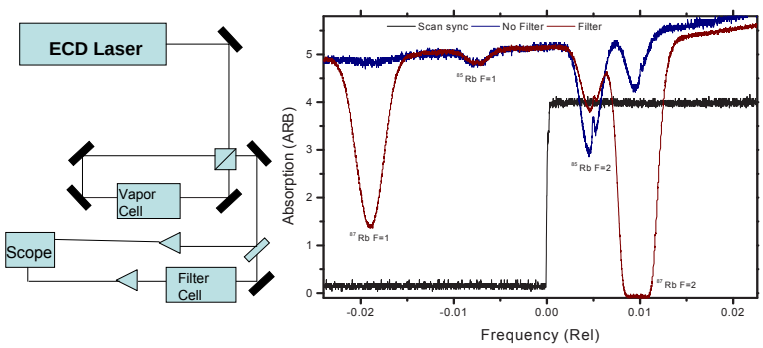

Figure 8. Left hand side show the optical set up for Doppler free spectroscopy of ^{85}Rb and ^{87}Rb with and without the isotope enriched filte . The Right hand side shows the results. The blue trace shows that the spectra with out the filte . The red trace is with the filte . The filte removes all the leakage light on the main ^{87}Rb.

3.4 Results

The main experiment of observing kinematically cooled ^{85}Rb via collisions with ^{87}Rb was performed using the florescenc imaging described above. A ^{87}Rb MOT with a density of $\sim 10^{10}$ atoms was loaded from a Zeeman decelerated beam. The atomic oven supplying the ^{87}Rb contains Rubidium metal with the natural isotope mixture of 72% ^{85}Rb and 28% ^{87}Rb. Thermal ^{85}Rb from the atomic oven enters the chamber and results in a small thermal background of ^{85}Rb. This background ^{85}Rb then undergoes collisions with the ^{87}Rb which can cool the ^{85}Rb down to below 1mK where the ^{85}Rb will be contained in the MOT magnetic field To detect the ^{85}Rb, the ^{87}Rb trapping lasers are acoustically switched off in 1μs and a set of near resonance ^{85}Rb lasers are switch on. The florescenc is monitored through our atomic filte imaging system, and the light is collected with either a photo-multiplier or a charged coupled device camera.

The initial data looks quite promising. We see an increase in the ^{85}Rb signal over the background when the ^{87}Rb is initially present. However, in performing the data analysis we discovered that there can be contributions to the signal, such as off-resonance light scattering, which could be contributing or occluding the actual data. For this reason we present no actual data here. To include such preliminary data in this discussion is improper, as the results and interpretation may potentially be incorrect.

The second set of experiments is focused on understanding and proving the basic physics behind the experimental techniques developed in the LDRD. The Kinematic cooling technique, as related to the Micro-Kelvin cold molecule LDRD, relies on kinetic energy transfer from elastic collisions to redistribute energy between the collision partners such that one particle is left with near zero kinetic energy in the laboratory frame while the collision partner carries away all the kinetic energy. The goal of this set of experiments is to show that (near) mass-resonant elastic collisions can kinematically cool particles from supersonic velocities to near stationary in the laboratory frame.

As the test particle we chose Krypton atoms. Krypton is a gas that can be easily purchased, produces supersonic molecular beams, has a similar mass (84 amu) to Rubidium, and is easily detected using (2+1) resonance enhanced multi-photon ionization, near 212nm. We generated two beams of neat Kr, and collided them at 90° in a vacuum chamber, a laser set 1.5mm above the collision plane then ionizes the atoms. The laser beam is translated out of the collision plane to help distinguish between the cold scattered atoms and the parent atomic beams which have several orders on magnitude more density. Once ionized, the Krypton ions are collected using the velocity-mapped ion-imaging technique allowing us to distinguish between hot (high velocity) and cold (low velocity) atoms.

Figure 9 shows a series of time delayed velocity-mapped ion-images. Knowing the times and the distances allows us to estimate the temperature, which is in the milli-Kelvin range. The results clearly show that we are able to use single near mass degenerate elastic collisions to cool particles from supersonic (hyper-thermal) velocities (over 400m/s for Kr) down into the milli-Kelvin regime. Modelling of the data shows a significan number of atoms have post collision velocities below 4 m/s.

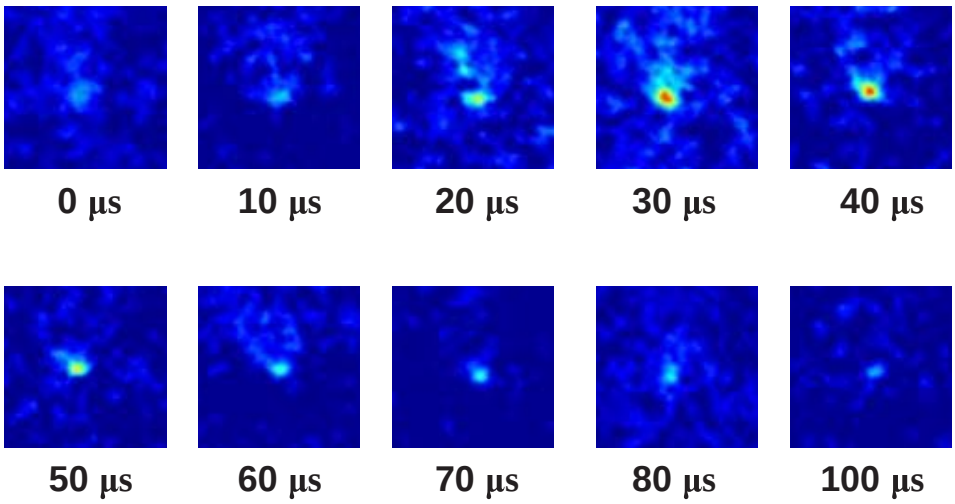

Figure 9. Time delayed images of the cold Krypton atoms produces via elastic kinematic cooling. The cold Krypton fit to temperatures in the milli-Kelvin range, and demonstrates the physics behind kinematic cooling with elastic collision between similar mass particles.

Thus, we have demonstrated that the underlying physics behind the cooling technique for producing micro-Kelvin cold molecules works, and further experiments are needed to clarify the origins of the potential ^{85}Rb signal. These experiments are currently on going.

4 Conclusion

The Micro-Kelvin cold molecule LDRD proposed a novel variation of the kinematic cooling technique for generating samples of micro-Kelvin temperature molecules. Because the kinematic technique only relies on a single collision, molecules can be cooled even with an unfavorable ratio of elastic to inelastic collision cross-sections. We have shown that for a test system of $D^{85}Rb$ scattering off of ^{87}Rb, we can expect to produce $\sim 10^5$ milli-Kelvin molecules-cm^{-3} from a room temperature vapor, and even larger numbers if the initial molecular velocities are reduced. Initial results are currently inconclusive. We have demonstrated the underlying physics behind this technique by demonstrating that elastic collisions between Krypton atoms in supersonic beams can be use to kinematically cool the atoms to the milli-Kelvin regime. Similarly, the models predict for $D^{81}Br$ scattering from ^{85}Rb we may be able to produce $\sim 10^6$ molecules-cm^{-3} at milli-Kelvin temperatures assuming we can start with $D^{81}Br$ cooled to liquid nitrogen temperatures. As the milli-Kelvin molecules are trapped coincident with the micro-Kelvin temperature atoms, further collisional cooling may occur, offering the possibility of samples of ground state molecules at micro-Kelvin temperatures.

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