SANDIA REPORT

SAND2009-6026
Unlimited Release
Printed September 2009

Molecular Fountain

David W. Chandler, Kevin E. Strecker

Prepared by
Sandia National Laboratories
Albuquerque, New Mexico 87185 and Livermore, California 94550

Sandia is a multiprogram laboratory operated by Sandia Corporation,
a Lockheed Martin Company, for the United States Department of Energy's
National Nuclear Security Administration under Contract DE-AC04-94-AL85000.

Approved for public release; further dissemination unlimited.

Sandia National Laboratories

Issued by Sandia National Laboratories, operated for the United States Department of Energy by Sandia Corporation.

NOTICE: This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government, nor any agency thereof, nor any of their employees, nor any of their contractors, subcontractors, or their employees, make any warranty, express or implied, or assume any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represent that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government, any agency thereof, or any of their contractors or subcontractors. The views and opinions expressed herein do not necessarily state or reflect those of the United States Government, any agency thereof, or any of their contractors.

Printed in the United States of America. This report has been reproduced directly from the best available copy.

Available to DOE and DOE contractors from
U.S. Department of Energy
Office of Scientific and Technical Information
P.O. Box 62
Oak Ridge, TN 37831

Telephone: (865) 576-8401
Facsimile: (865) 576-5728
E-Mail: reports@adonis.osti.gov
Online ordering: http://www.osti.gov/bridge

Available to the public from
U.S. Department of Commerce
National Technical Information Service
5285 Port Royal Rd
Springfield, VA 22161

Telephone: (800) 553-6847
Facsimile: (703) 605-6900
E-Mail: orders@ntis.fedworld.gov
Online ordering: http://www.ntis.gov/help/ordermethods.asp?loc=7-4-0#online

SAND2009-6026
Unlimited Release
Printed September 2009

Molecular Fountain

David W. Chandler
Chemical Sciences
Sandia National Laboratories
P.O. Box 969 MS 9055
Livermore, CA 94551-0969
chand@sandia.gov

Kevin E. Strecker
Combustion Chemistry Department
Sandia National Laboratories
P.O. Box 969 MS 9055
Livermore, CA 94551-0969
kstreck@sandia.gov

Abstract

A molecular fountain directs slowly moving molecules against gravity to further slow them to translational energies that they can be trapped and studied. If the molecules are initially slow enough they will return some time later to the position from which they were launched. Because this round trip time can be on the order of a second a single molecule can be observed for times sufficien to perform Hz level spectroscopy. The goal of this LDRD proposal was to construct a novel Molecular Fountain apparatus capable of producing dilute samples of molecules at near zero temperatures in well-defined user-selectable, quantum states. The slowly moving molecules used in this research are produced by the previously developed Kinematic Cooling technique, which uses a crossed atomic and molecular beam apparatus to generate single rotational level molecular samples moving slowly in the laboratory reference frame. The Kinematic Cooling technique produces cold molecules from a supersonic molecular beam via single collisions with a supersonic atomic beam. A single collision of an atom with a molecule occurring at the correct energy and relative velocity can cause a small fraction of the molecules to move very slowly vertically against gravity in the laboratory. These slowly moving molecules are captured by an electrostatic hexapole guiding fiel that both orients and focuses the molecules. The molecules are focused into the ionization region of a time-of-fligh mass spectrometer and are ionized by laser radiation.

The new molecular fountain apparatus was built utilizing a new design for molecular beam apparatus that has allowed us to miniaturize the apparatus. This new design minimizes the volumes and surface area of the machine allowing smaller pumps to maintain the necessary background pressures needed for these experiments.

Contents

Summary 6
1 Inroduction 7
2 Review of ultracold molecule fiel 9
3 Kinematic cooling 11
4 Molecular Fountain Apparatus 15
5 Conclusion 18
References 19

Figures

1 Time delayed images of cold NO(X)j=7.5. 11
2 Normalized production and decay of the NO(X)j=7.5 compared with the NO(X)j=10.5. 13
3 Images of inelastically-scattered ND_3 products in low-lying quantum states. 14
4 Representation of scattering process that produces a slowly moving sample of molecules moving vertically in the laboratory. 15
5 Schematic drawing of the experiment. 16
6 Photograph of the molecular fountain. 17

Summary

A device was built that guides and focuses slowly moving molecules. The slowly moving molecules are formed by a collision between a molecule seeded into a supersonic beam having a collision with an atom in a separate supersonic beam. A small fraction of the collisions will create molecules that are moving slowly in the lab opposite in direction to the force of gravity. These molecules enter a electrostatic hexapole fiel that focuses them as they travel up, against gravity, in the laboratory. The hexapole fiel acts as a lens for molecules that populate low fiel seeking stark states. These molecules will be focused at the exit of the hexapole that coincides with the ionization region of a time of fligh mass spectrometer. A laser beam intersects the focused molecules and after the molecule absorbs several photons the molecule is ionized. The ions are then projected onto a time and position sensitive ion detector where their arrival time and location are recorded. This apparatus when fully developed, will be a new scientifi capability at Sandia.

1 Inroduction

For the last ten years groups around the world have been searching for ways to cool molecules. The cooling of atoms to ultra cold temperatures has resulted in spectacular discoveries. The realization and study of new states of matter like Bose Einstein Condensates, degenerate Fermi gasses and BCS (Bardeen, Cooper Schrieffer) fluid are now "routine". In addition to these new states of matter that have been formed, cooling atoms to microKelvin temperatures, and below, has enabled ultra-high resolution spectroscopy studies utilizing an apparatus known as an atomic fountain. The atomic fountain utilizes six counter propagating laser beams that exert pressure on the atoms bringing them to a near stand still (a few mm/s velocity) at the intersection region of the laser beams. This creates a sample of atoms with a temperature in the microKelvin range (a few millionths of a degree Kelvin). They are then launched up against gravity utilizing one of the laser beams to push them. As they move up they are excited in a microwave cavity both as they move up and as they return to the position of origin. The influenc of the microwaves is monitored by laser-induced fluorescenc of the sample after the second pass through the microwave cavity. In this manner the exact energy at which the atom absorbs microwave radiation is determined. The frequency of that light (9,192,631,770 Hz for Cesium) is used to defin the second.

If a similar apparatus can be built for molecules very high-resolution spectroscopy can be performed on molecular samples. Two problems of importance that can be addressed by ultra-high spectroscopy on molecules are; 1) to determine the magnitude of the dipole of the electron and 2) to determine the mass of the hydrogen atom. The dipole moment of the electron is a measure of the average distance that the charge resides from the center-of-mass of the electron. As Ed Hinds of Imperial College London writes " The dipole moment is parallel to the direction which the electron spins because any other component of charge distribution would average to zero by the motion of the spin. This implies that the product of an electron's spin and electric dipole moment is an intrinsic property of the electron. Since the spin reverses if time runs backwards, the dipole moment must be zero if the electron is not to violate 'time reversal symmetry' - a postulate of the Standard Model. Any other value would mean the Standard Model needs to be revised." The dipole moment of the electron can be determined from ultrahigh resolution spectroscopy on a molecule containing a very heavy nucleus, such as YbF [22]. Wim Ubach of the Free University of Amsterdam is attempting to make a molecular fountain of ammonia, NH_3. By measuring with ultra-high resolution spectroscopy the umbrella mode of the molecule one can deduce the mass of the hydrogen atom. By comparing the measured mass of hydrogen with that deduced from astronomical measurements one can determine if the mass is constant with time. A variation of a constant of nature implies that the laws of nature are subject to change [24].

We have built a molecular fountain by directly scattering molecules in a direction anti-parallel to the force of gravity. We collect these molecules with an electrostatic hexapole guiding field This hexapole fiel separates the scattered molecules from the background molecules formed in the scattering process. The hexapole not only separates out the molecules of interest it concentrate them and selects only those that populate quantum states that have low-fiel seeking Stark behavior. The molecules that emerge from the hexapole fiel are extremely well characterized and this state preparation will be useful for photochemistry experiments.

Here we will describe a molecular fountain apparatus that takes advantage of Kinematic cooling physics to cool molecules. The vertically scattered molecules are focused into the ionization region of a time-of-fligh mass spectrometer. They are laser ionized and detected by velocity mapped Ion Imaging technique. We have recently developed a new design for assembling molecular beam apparatus that will be adapted to form the molecular fountain. The apparatus features two doubly skimmed and velocity selected molecular beams that cross directly below the hexapole guiding field By picking the molecule and atom for scattering we can determine the speed and quantum state of the vertically scattered molecules. For instance if we scatter NO molecules from Ar atoms then the vertically scattered slow molecules will be NO in the j=6.5 quantum state. If we scatter NH_3 from Ne then the vertically scattered slow NH_3 molecule will be in the j=1 quantum state.

2 Review of ultracold molecule field

The cooling of molecules to sub Kelvin temperatures promises to have great impact in the fiel of chemistry and physics. At these temperatures molecules are slowly moving in the laboratory reference frame and offer the possibility of long observation times needed for ultrahigh resolution spectroscopy. Their low kinetic energy also allows one to trap them with strong external electric, magnetic or optical field allowing for the study of collisional events as well making them attractive targets for evaporative cooling to lower temperatures. Additionally, their low momentum implies a large de Broglie wavelength that allows the molecules to exhibit their wave nature as well as their particle nature.

A distinction needs to be made between the "cold" and "ultracold" sub Kelvin temperature regimes as these terms have particular meaning to certain sub field of physics and chemistry. There is no rigorous definitio for the "cold" regime. "Cold" is typically taken to be around the temperature at which the thermal de Broglie wavelength is becoming larger than the classical size of the particle. However, the cold regime is also used to refer to any range of low temperatures at which new physical phenomena, not accessible by standard techniques, can be explored. One useful definitio for the cold regime is to have particles at sufficientl low temperature/energy such that they can readily be manipulated, confined or stored using external fields For many new chemistry and physics experiments the "cold" regime is all that is needed. The ability to trap, store and manipulate along with the extended observation times allows for ultrahigh resolution spectroscopy that, for example, may allow a direct counting of the density of states of the molecules[5] and precision measurements of fundamental physical constants[16, 24, 35, 20]. Confinin and orientation of the molecules prior to photo-dissociation reduces the spatial averaging associated with a randomly oriented molecule thereby yielding new details in the observed angular distributions of the photofragments that can reveal the underlying dynamics of the photo-dissociation process [43]. Further, the wave nature of the particles can be exploited through the molecular analog of atom optics. The increased thermal de Broglie wavelength makes it possible to perform unique interferometric studies with molecules, such as the separation of mixtures of clusters by diffraction through a transmission grating.[33, 29, 42, 26, 10]

Beyond the ability to do physical chemistry type of experiments there are many high precision spectroscopy based physics experiments that would greatly benefi from such samples. The vibrational and rotational transition frequencies in molecules can be used for experiments searching for a time-dependence of the electron-to-proton and nucleon-to-nucleon mass ratios [16]. In a recent paper it was described how the worlds most sensitive interferometer, capable of the most precise measurement of gravity waves, can be made from coherent excitation of ro-vibrational states of trapped cold molecules[46].

In contrast to the cold regime, the "ultracold" regime is more clearly defined The term ultracold refers to a sample in which the thermal de Broglie wavelength is on the same order as the average inter-particle separation, such that $\lambda_{db} > n^{-\frac{1}{3}}$. Where, n, is the particle density. The physical meaning of this relationship is that due to their quantum size the particles are beginning to spatially overlap. The particle interactions are no longer classical hard-sphere-like interactions but are now predominantly wavelike, and are governed by their respective quantum statistics. The

cooling of atoms to ultracold temperatures, where the quantum nature of the gas dominate their interactions, has resulted in spectacular discoveries, such as; the realization and study of new states of matter like Bose Einstein Condensates [1, 2, 9], degenerate Fermi gasses [12, 41, 11, 18] and BCS [7, 4, 19] (Bardeen, Cooper Schrieffer) fluids In addition to forming these new states of matter, cooling atoms to microKelvin temperatures, and below, has enabled both ultrahigh resolution spectroscopy studies [45, 6, 22] and the extraction of information about the collisional dynamics of atoms and their interactions (for example; shape resonances[36, 47], Feschbach resonances [21, 28, 23, 44] and Efim v states [37, 8, 3]). In addition the purely quantum behavior like matter-wave lasers [31] and matter-wave solitons [40], and the ability to study complex super-conducting behavior in nearly ideal dilute quantum gases [11, 19, 32, 48] have been studied at ultracold temperatures. All of these systems have molecular analogs and the specifi behavior, scattering lengths, location of shape and Feshbach resonances depend strongly on the shape and structure of the potential energy surface on which the particles interact. The added complexity found in molecules including, permanent dipoles and quadrapole moments, complex rotational and vibrational structure and chemistry, offer the possibility of rich areas of investigation in areas such as spectroscopy and collision dynamics. However, the field of cold and ultracold molecule studies had remained generally unexplored due to the complexity of making cold and ultracold samples of molecules.

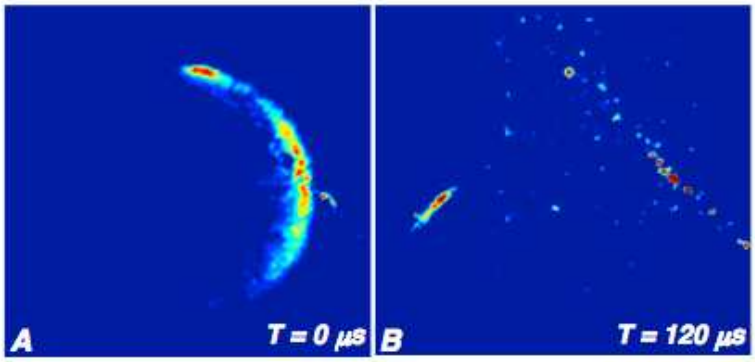

Figure 1. Image on the left is a velocity-mapped ion-image of the NO(X)j=7.5 at the peak of the scattering between the NO and Ar. All velocity produced during the collision process are visible. The image on the right is taken after a 120 μs delay. We observe that all the molecules with velocities other then the near zero have fl w out of our observation region. Trace amounts of NO are also visible, which originate from the effusion of gas from the source regions.

3 Kinematic cooling

Kinematic Cooling is a unique approach to the production of ultracold molecules, slowing of a molecule by a single collision with an appropriate atom bringing a subset of the molecules to rest in the laboratory reference frame. We have developed the Kinematic cooling technique for the "routine" production of samples of millikelvin temperature molecules. The crossed molecular beam scattering of a molecular beam from an atomic beam at 90° geometry can create a sample of molecules moving very slowly in the laboratory reference frame. By picking the masses of the particles and the velocity of the molecule one can dictate that molecules populating a particular quantum state come to rest in the laboratory. We have produced measurable amounts of cold (milliKelvin temperature range) molecules (NO, NH_3, HCl) using this technique. Recently we reported on a redesign of our molecular beam apparatus has allowed us to not only produce kinematicaly cooled molecules but to also extract them from the high density molecular and atomic beams. This is a major advancement in the kinematic cooling technique as it allows us to now trap the cold molecules with external electric and magnetic fields Additionally it implies that slow vertically scattered molecules can escape the molecular beams and be injected into the molecular fountain's hexapole guide.

The major obstacle in kinematic cooling has been that only a small number of the total collisions results in the formation of cold molecules, and the cold molecules are necessarily formed in the interaction volume of the atomic and molecular beams. As a result, cold molecules have a

high probability of secondary glancing collisions with the atomic and molecular beams that re-heat them. In the past year we have reported[38] on our success at reducing the destructive re-heating collisions for the cold molecules such that our observation time is consistent with being limited by diffusion of the cold molecules from our interaction region and not secondary collisions. Under these conditions we are able to observe unconfined kinematically cooled, ground vibrational Nitric Oxide (NO), in the j=7.5 rotational state for over 150 μs. An upper limit for the fina average velocity is estimated from the diffusion time of the cold molecules from the observation region define by the intersection of the atomic and molecular beams with the detection laser. The diffusion of the cold molecules is modeled using a three-dimensional Monte-Carlo simulation to account for the size of the interaction region and laser beam overlap. These simulations are consistent with the data and give an average fina velocity of approximately, 4.5 m/s for the NO(X)j=7.5. There is no reason to believe that the kinematically cooled molecules are in thermal equilibrium, however, as a matter of convention we use the relationship $1/2m\ v^2 = k\ T$ to relate the velocity to a temperature. Here v is the average velocity, m is the mass, k is the Boltzmann constant, and T is the temperature. For NO, with a 4.5 m/s fina average velocity, one calculates a temperature of approximately 35 mK. This represents a reduction in the observed "temperature" of over an order of magnitude from our previous report [3]. We are now able to observe a single sample of cold molecules in a fiel free region for up to 150 μs which allows for the possibility of high resolution, $\sim$10 kHz, molecular spectroscopy. If these molecules can be injected into a molecular fountain with a round trip time of one second then Hz resolution spectroscopy is possible. This is a four order of magnitude improvement. In general spectroscopy in molecular beams is limited to MHz (10^6 Hz) resolution due to fly-ou time of the molecules from the laser beam.

The important result is graphically summarized in figur 2. We have, for the firs time, been able to establish a set of experimental conditions such that the cold molecules produced via Kinematic cooling are temporally separated from the parent atomic and molecular beams. This conclusion is supported by our observations of the quantum state dependence of the bi-modal decays for both the NO(X) j=7.5 and j=10.5 rotational states. The temporally separated cold molecules can now serve as a starting point for several new experiments including trapping and storing molecules allowing for cold collisional studies and high resolution molecular spectroscopy. This would be the firs time the products of a crossed molecular beam scattering experiment were used as the reagents for a new experiment instead of simply detected. We have recently reported [27] on utilizing this technique for the cooling of ammonia molecules in collision with Ne. The ND_3 + Ne system as this should result in even colder molecules than the NO + Ar system due to the closer mass match between the atom and molecule. In Figure 3 we display some images we have obtained from ammonia scattering from neon. The bright spot on the top of the images of figur 3 are the cold molecules.

The images of the rotational states which are expected to yield the coldest molecules are shown in Figure 3. For these states, range from 14.48 cm^{-1} for the J = 2, K = 2 state, up to 51.42 cm^{-1} for the J = 3, K = 1 state. Although scattering tends toward relatively low angles for these low quantum states, in every case there is enough scattering around 90 degrees to produce a clear build-up of cold molecules. To determine the velocities of the scattered products, the data are fi in the same manner as above. A section through the scattering ring at 95 degrees is then taken, and the velocity distribution is fi to a Gaussian to determine the center velocity (the cold molecules

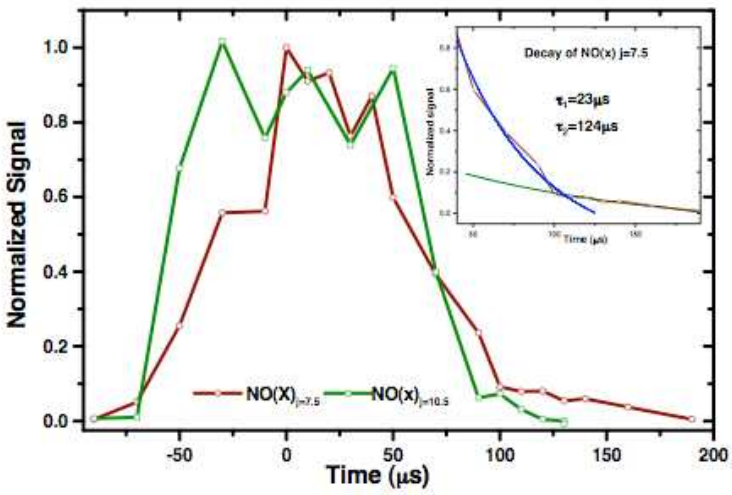

Figure 2. Normalized production and decay of the NO(X)j=7.5 compared with the NO(X)j=10.5 . The NO(X)j=10.5 data is given by the marked with the square points and colored green while the NO(X)j=7.5 is marked with the circular points and colored red. We attribute the loss between 50 and 70 μs to secondary collisions with the parent atomic and molecular beams. The decay from 75μs on is attributed to the cold molecules flyin out of the probing region. In this long time region, after 75μs, we observe a 19 $\pm$ 8μs decay for the NO(X)j=10.5 while we observe a longer, 124 $\pm$ 4μs, decay for the NO(X)j=7.5. The errors in the decays are found by minimizing χ^2 value of the exponential fits

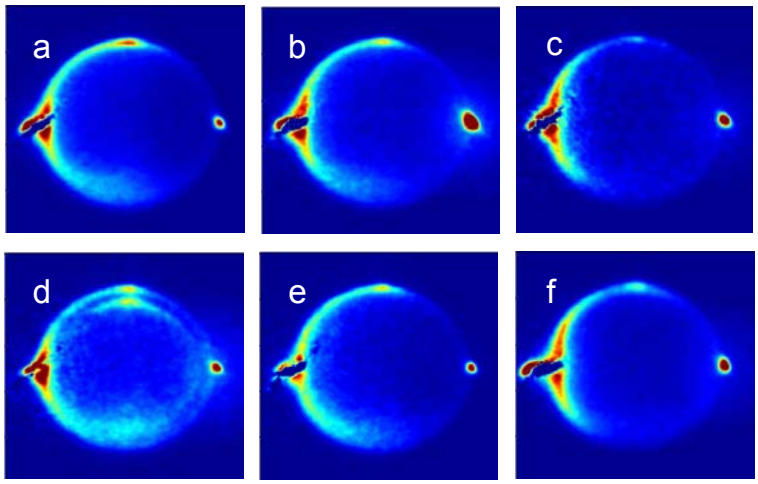

Figure 3. Images of inelastically-scattered ND_3 products in low-lying quantum states. (a) J = 2, K = 0; (b) J = 2, K = 1; (c) J = 2, K = 2; (d) J = 3, K =1; (e) J = 3, K = 2; (f) J = 3, K = 3. A second ring, caused by scattering into a higher rotational level, is found in the interior of (d), and is observed due to a blended optical transition.

are not centered at zero velocity in the laboratory reference frame) and the velocity spread. The velocities of the centers of the distributions associated with the cold molecules are measured to be 20-30 m/s (with an uncertainty of 10 m/s), and the velocity spreads of the cold molecules are typically found to be 20 m/s FWHM. The calculations indicate that the molecules are even slower, and predict center velocities ranging from 7.5 m/s up to 22.8 m/s with velocity spreads of 1 m/s or less. The calculations predict much lower temperatures: 70 - 880 mK in the laboratory frame, and 80 μK - 1.1 mK in the moving frame. Due to imperfections in our ability to image such slow moving molecules along with the inherent problems associated with extracting velocities from two-dimensional projections (the images of figur 3) our measurements of velocities and temperatures represent upper limits. Despite this we see that we should be able to trap several states of the ammonia molecule with electrostatic traps.

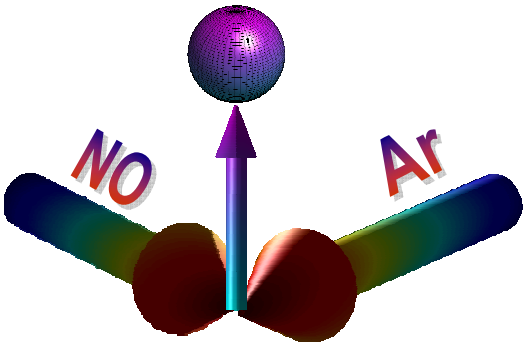

Figure 4. Representation of scattering process that produces a slowly moving sample of molecules moving vertically in the laboratory.

4 Molecular Fountain Apparatus

The concept of the Kinematic Cooling to produce a sample of slowly moving vertically scattered molecules is shown in figur 4. One supersonic beam of molecules collides with a supersonic beam of atoms. The result is a sample of molecules moving vertically in the laboratory. These molecules will be injected into a hexapole guiding fiel for focusing and separation.

The slowly moving molecular sample entering the bottom of the hexapole guiding fiel will be focused into the ionization region of a time-of-fligh ass spectrometer. This is schematically shown in figur 5. The ionization region of the time-of-fligh mass spectrometer consists of velocity-mapped Ion Imaging optics. These focus the laser formed ions onto a position sensitive ion detector that is backed by a phosphor screen. The ions are then detected by recording the light pulses that come from the phosphor, either on a CCD camera or a photomultiplier tube. The ion optics consist of a repeller plate with a positive voltage, to push the positively charged ions toward the detector, a Extractor plate, that acts as a lens and focuses the ions onto the imaging detector, and a ground plate that serves as an entrance to the time-of-fligh tube. A photograph of the hexapole and the ion optics is shown in Figure 6.

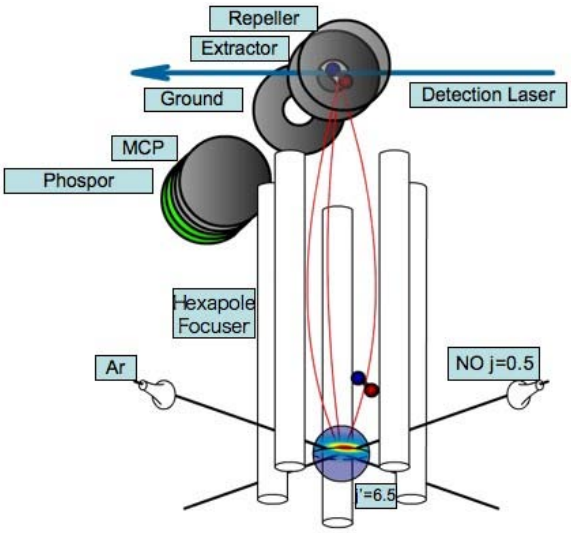

Figure 5. Schematic drawing of the experiment. A molecular (NO) and atomic (Ar) beam cross at the bottom of a hexapole guiding filed Some molecules are scattered into the hexapole and focused into the ionization region of a mass spectrometer.

Figure 6. Photograph (upside down) of the electrostatic hexapole guiding file for dipolar molecules with low fiel seeking Stark states. Molecules entering the bottom of the hexapole will be focused into the ionization region of a time-of-fligh mass spectrometer.

5 Conclusion

A prototype molecular fountain based on Kinematic Cooling of molecules has been built. The apparatus consist of crossed molecular and atomic beams that create slow vertically scattered molecules. These are focused into a electrostatic hexapole fiel for separating and focusing them. First experiments are under way utilizing NO scattered from Ar and NH_3 / ND_3 from Ne.

References

[1] M. H. Anderson, J. R. Ensher, M. R. Matthews, C. E. Wieman, and E. A. Cornell. Observation of Bose-Einstein condensation in a dilute atomic vapor. *Science*, 269:198, 1995.

[2] M R Andrews, M-O Mewes, N J van Druten, D S Durfee, D M Kurn, and W Ketterle. Direct, nondestructive observation of a Bose condensate. *Science*, 273:84, 1996.

[3] I. Baccarelli, G. Delgado-Barrio, F.A. Gianturco, T. Gonzalez-Lezana, S. Miret-Artes, and P. Villarreal. Searching for efim v states in triatomic systems: The case of $LiHe_2$. *European physics Letters*, 50:567, 2000.

[4] M. Bartenstein, A. Altmeyer, S. Riedl, S. Jochim, C. Chin, JH Denschlag, and R. Grimm. Collective excitations of a degenerate gas at the BEC-BCS crossover. *Physical Review Letters*, 92:203201, 2004.

[5] C. A. Biesheuvel, J. Bulthuis, M. H. M. Janssen, S. Stolte, and J. G. Snijders. High-resolution laser spectroscopy of no[sub 2] just above the x-tilde[sup 2]a[sub 1]–[a-tilde][sup 2]b[sub 2] conical intersection: Transitions of k[sub -] = 1 stacks. *The Journal of Chemical Physics*, 112(8):3633–3642, 2000.

[6] J.J. Bollinger, W.M. Itano, D.J. Wineland, and D.J. Heinzen. Optimal frequency measurements with maximally correlated states. *Physical Review A*, 54:R4649, 1996.

[7] T. Bourdel, L. Khaykovich, J. Cubizolles, J. Zhang, F. Chevy, M. Teichmann, L. Tarruell, SJJMF Kokkelmans, and C. Salomon. Experimental study of the bec-bcs crossover region in lithium 6. *Physical Review Letters*, 93:050401, 2004.

[8] E. Braaten, H.W. Hammer, and M. Kusunoki. Efim v states in a bose-einstein condensate near a Feshbach resonance. *Physical Review Letters*, 90:170402, 2003.

[9] C. C. Bradley, C. A. Sackett, J. J. Tollett, and R. G. Hulet. Evidence of Bose-Einstein condensation in an atomic gas with attractive interactions. *Physical Review Letters*, 75:1687–1690, 1995.

[10] R. Bruhl, A. Kalinin, O. Kornilov, J.P. Toennies, G.C. Hegerfeldt, and M. Stoll. Matter wave diffraction from an inclined transmission grating: Searching for the elusive He-4 trimer Efim v state. *Physical Review Letters*, 95:063002, 2005.

[11] C. Chin, M. Bartenstein, A. Altmeyer, S. Riedl, S. Jochim, J. Hecker Denschlag, and R. Grimm. Observation of the pairing gap in a strongly interacting Fermi gas. *Science*, 305:1128, 2004.

[12] B. DeMarco and D. S. Jin. Onset of Fermi degeneracy in a trapped atomic gas. *Science*, 285:1703, 1999.

[13] MS Elioff, JJ Valentini, and DW Chandler. Subkelvin cooling NO molecules via ”billiard-like” collisions with argon. *Science*, 302:1940–1943, 2003.

[14] MS Elioff, JJ Valentini, and DW Chandler. Formation of NO(j '=7.5) molecules with sub-kelvin translational energy via molecular beam collisions with argon using the technique of molecular cooling by inelastic collisional energy-transfer. *European Physical Journal D*, 31:385–393, 2004.

[15] M. Faubel, K.-H. Kohl, J.P. Toennies, Kwong Tin Tang, and Yat Yan Yung. The He-N/sub 2/ anisotropic van der Waals potentials: test of a simple model using state-to-state differential scattering cross-sections. *Faraday Discussions of the Chemical Society*, (73):205–20, 1982 1982.

[16] U. Frohlich, B. Roth, P. Antonini, C. Lammerzahl, A. Wicht, and S. Schiller. Ultracold trapped molecules: novel systems for tests of the time-independence of the electron-to-proton mass ratio. In S.G. Karshenboim and E. Peik, editors, *Astrophysics, clocks and fundamental constants (Lecture Notes in Phys. Vol.648)*, pages 297–307. 2004 2004.

[17] A Gijsbertsen, H Linnartz, G Rus, AE Wiskerke, S Stolte, DW Chandler, and J Klos. Differential cross sections for collisions of hexapole state-selected NO with He. *Journal of Chemical Physics*, 123:224305–1–14, 2005.

[18] M. Greiner, C.A. Regal, J.T. Stewart, and D.S. Jin. Probing pair-correlated fermionic atoms through correlations in atom shot noise. *Physical Review Letters*, 94(11):110401/1–4, 25 March 2005.

[19] Kevin E. Strecker Guthrie B. Partridge and, Ramsey I. Kamar, Michael W. Jack, and Randall G. Hulet. Molecular probe of pairing in the BEC-BCS crossover. *Physical Review Letters*, 95:020404, 2005.

[20] EA Hinds. Testing time reversal symmetry using molecules. *PHYSICA SCRIPTA*, T70:34–41, 1997.

[21] M. Houbiers, H. T. C. Stoof, W. I. McAlexander, and R. G. Hulet. Elastic and inelastic collisions of ^{6}Li atoms in magnetic and optical traps. *Physical Review A*, 57:R1497, 1998.

[22] JJ Hudson, BE Sauer, MR Tarbut, and EA Hinds. Measurement of the electron electric dipole moment using YbF molecules. *Physical Review Letters*, 89:023003, 2002.

[23] S. Inouye, M. R. Andrews, J. Stenger, H.-J. Miesner, D. M. Stamper-Kurn, and W. Ketterle. Observation of Feshbach resonances in a Bose-Einstein condensate. *Nature*, 392:151, 1998.

[24] T. I. Ivanov, M. Roudjane, M. O. Vieitez, C. A. de Lange, W.-U L. Tchang-Brillet, and W. Ubachs. Hd as a probe for detecting mass variation on a cosmological time scale. *Physical Review Letters*, 100(9):093007, 2008.

[25] Paul Jansen and David W. Chandler Kevin E. Strecker. Kinematic production of isolated millikelvin molecules. *Review of Scientific Instruments*, 80:083105, 2009.

[26] A. Kalinin, O. Kornilov, W. Schollkopf, and J.P. Toennies. Observation of mixed fermionic-bosonic helium clusters by transmission grating diffraction. *Physical Review Letters*, 95:113402, 2005.

[27] JJ Kay, SYT van de Meerakker, KE Strecker, and DW Chandler. Production of cold ND_3 by kinematic cooling. *Faraday Discussions*, 142:143–153, 2009.

[28] A.J. Kerman, C. Chin, V. Vuletic, S. Chu, P.J. Leo, C.J. Williams, and P.S. Julienne. Determination of Cs-Cs interaction parameters using feshbach spectroscopy. *Comptes Rendus de l'Academie des Sciences, Serie IV (Physique, Astrophysique)*, 2:633, 2001.

[29] D.A. Lapshin, V.I. Balykin, and V.S. Letokhov. Diffraction of atoms by an evanescent wave grating formed by a periodic surface structure. *Laser Physics*, 7:361, 1997.

[30] P McCabe and JNL Connor. Nearside farside analysis of differential cross sections: Diffraction and rainbow scattering in atom-atom and atom-molecule rotationally inelastic sudden collisions. *JOURNAL OF CHEMICAL PHYSICS*, 104(6):2297–2311, FEB 8 1996.

[31] M.-O. Mewes, M. R. Andrews, D. M. Kurn, D. S. Durfee, C. G. Townsend, and W. Ketterle. Output coupler for Bose-Einstein condensed atoms. *Physical Review Letters*, 78(4):582, 1997.

[32] GB Partridge, WH Li, RI Kamar, YA Liao, and RG Hulet. Pairing and phase separation in a polarized fermi gas. *SCIENCE*, 311(5760):503–505, JAN 27 2006.

[33] T.M. Roach. Atom wave diffraction in an accelerating potential. *Journal of Physics B*, 37:3551, 2004.

[34] W.G. Roeterdink, K.E. Strecker, C.C. Hayden, M.H.M. Janssen, and D.W. Chandler. Imaging the rotationally state-selected NO(A,n) product from the predissociation of the a state of the NO-Ar van der Waals cluster. *Journal of Chemical Physics*, 130(13):134305 (8 pp.), 7 April 2009.

[35] E. J. Salumbides, D. Bailly, A. Khramov, A. L. Wolf, K. S. E. Eikema, M. Vervloet, and W. Ubachs. Improved laboratory values of the h_2 lyman and werner lines for constraining time variation of the proton-to-electron mass ratio. *Physical Review Letters*, 101(22):223001, 2008.

[36] R. Stock, I.H. Deutsch, and E.L. Bolda. Quantum state control via trap-induced shape resonance in ultracold atomic collisions. *Physical Review Letters*, 91:183201, 2003.

[37] M. Stoll and T. Kohler. Production of three-body Efim v molecules in an optical lattice. *Physical Review A*, 72:22714, 2005.

[38] Kevin E. Strecker and David W. Chandler. Kinematic production of isolated millikelvin molecules. *Physical Review A*, 78(6):063406, 2008.

[39] Kevin E. Strecker and David W. Chandler. *Low Temperatures and cold molecules*. Imerial College Press, Editor; Ian W. Smith, UK, 1st edition, 2008.

[40] Kevin E. Strecker, Guthrie B. Partridge, Andrew G. Truscott, and Randall G. Hulet. Formation and propagation of matter-wave soliton trains. *Nature*, 417:150–153, 2002.

[41] A.G. Truscott, K.E. Strecker, W.I. McAlexander, G.B. Partridge, and R.G. Hulet. Sympathetic cooling of ^{6}Li atoms. *Science*, 10593181-1:3, 2001.

[42] J. Vale, B.V. Hall, D.C. Lau, M.P.A. Jones, J.A. Retter, and E.A. Hinds. Atom chips. *Europhys News*, 33, 2002.

[43] AJ van den Brom, TP Rakitzis, and MHM Janssen. Molecular frame properties from photodissociation of laboratory-oriented symmetric top and chiral molecules. *PHYSICA SCRIPTA*, 73(1):C83–C88, JAN 2006.

[44] T. Volz, S. Durr, N. Syassen, G. Rempe, E. Van Kempen, and S. Kokkelmans. Feshbach spectroscopy of a shape resonance. *Physical Review A*, 72:010704, 2005.

[45] J. Weiner, V. Bagnato, S. Zilio, and P. S. Julienne. Experiments and theory in cold and ultracold collisions. *Reviews of Modern Physics*, 71:1, 1999.

[46] Andreas Wicht, Claus Lammerzahl, Dennis Lorek, and Hansjorg Dittus. Rovibrational quantum interferometers and gravitational waves. *Physical Review A (Atomic, Molecular, and Optical Physics)*, 78(1):013610, 2008.

[47] C.J. Williams, E. Tiesinga, and P.S. Julienne. Hyperfin structure of the Na_2 O_g^- long range molecular state. *Phys Rev A*, 53:R1939, 1996.

[48] M.W. Zwierlein, J.R. Abo-Shaeer, A. Schirotzek, C.H. Schunck, and W. Ketterle. Vortices and superfluidit in a strongly interacting Fermi gas. *Nature*, 435:1047–1051, 2005.

Sandia National Laboratories