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14. ABSTRACT
This project was superseded and replaced by another ARO-funded project of the same name, which is still continuing. The goal of this project was to explore ultracold collisions and chemistry between atoms and ions, to enable a novel cooling method, proposed by our group, for producing ultracold molecular ions. During this effort period, we experimentally and theoretically studied ultracold chemistry and collision in two important systems Ca + Yb+ and Ca + Ba+. As a result of this work, we have gained a clearer understanding of both the fundamental physics important for these collisions and the requirements for producing stable, long-lived ultracold ion-neutral

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Report Title

Final Report for Understanding Molecular-Ion Neutral Atom Collisions for the Production of Ultracold Molecular Ions

ABSTRACT

This project was superseded and replaced by another ARO-funded project of the same name, which is still continuing. The goal of this project was to explore ultracold collisions and chemistry between atoms and ions, to enable a novel cooling method, proposed by our group, for producing ultracold molecular ions. During this effort period, we experimentally and theoretically studied ultracold chemistry and collision in two important systems Ca + Yb⁺ and Ca + Ba⁺. As a result of this work, we have gained a clearer understanding of both the fundamental physics important for these collisions and the requirements for producing stable, long-lived ultracold ion-neutral systems. Building on this work, we have recently performed a proof-of-principle experiment that has demonstrated sympathetic cooling of both the translational and vibrational motion of BaCl⁺ molecules through collisions with ultracold gases. The results of this experiment are very promising and indicate that the proposed cooling method is as efficient as expected. Work now has turned to optimizing the experimental system for the production of ultracold, ground-state molecular ions.

Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

| <u>Received</u> | <u>Paper</u> |
|-----------------|---|
| 01/15/2014 | 7.00 Kuang Chen, Scott T. Sullivan, Wade G. Rellergert, Eric R. Hudson. Measurement of the Coulomb Logarithm in a Radio-Frequency Paul Trap, Physical Review Letters, (04 2013): 0. doi: 10.1103/PhysRevLett.110.173003 |
| 01/15/2014 | 5.00 Wade G. Rellergert, Scott T. Sullivan, Steven J. Schowalter, Svetlana Kotochigova, Kuang Chen, Eric R. Hudson. Evidence for sympathetic vibrational cooling of translationally cold molecules, Nature, (03 2013): 0. doi: 10.1038/nature11937 |
| 01/15/2014 | 6.00 Wade G. Rellergert, Scott T. Sullivan, Svetlana Kotochigova, Eric R. Hudson. Role of Electronic Excitations in Ground-State-Forbidden Inelastic Collisions Between Ultracold Atoms and Ions, Physical Review Letters, (11 2012): 0. doi: 10.1103/PhysRevLett.109.223002 |
| 07/29/2011 | 1.00 Kuang Chen, Steven Schowalter, Svetlana Kotochigova, Alexander Petrov, Wade Rellergert, Scott Sullivan, Eric Hudson. Molecular-ion trap-depletion spectroscopy of BaCl ⁺ , Physical Review A, (3 2011): 30501. doi: 10.1103/PhysRevA.83.030501 |
| 07/29/2011 | 2.00 Scott T. Sullivan, Wade G. Rellergert, Svetlana Kotochigova, Kuang Chen, Steven J. Schowalter, Eric R. Hudson. Trapping molecular ions formed via photo-associative ionization of ultracold atoms, Physical Chemistry Chemical Physics, (07 2011): 1. doi: 10.1039/c1cp21205b |
| 08/21/2012 | 3.00 Wade Rellergert, Scott Sullivan, Svetlana Kotochigova, Alexander Petrov, Kuang Chen, Steven Schowalter, Eric Hudson. Measurement of a Large Chemical Reaction Rate between Ultracold Closed-Shell ⁴⁰ Ca Atoms and Open-Shell ¹⁷⁴ Yb ⁺ Ions Held in a Hybrid Atom-Ion Trap, Physical Review Letters, (12 2011): 0. doi: 10.1103/PhysRevLett.107.243201 |
| 08/21/2012 | 4.00 Steven J. Schowalter, Kuang Chen, Wade G. Rellergert, Scott T. Sullivan, Eric R. Hudson. An integrated ion trap and time-of-flight mass spectrometer for chemical and photo- reaction dynamics studies, Review of Scientific Instruments, (2012): 0. doi: 10.1063/1.3700216 |
| TOTAL: | 7 |

Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

Received Paper

TOTAL:

Number of Papers published in non peer-reviewed journals:

(c) Presentations

1. S. Schowalter, C. Schneider, W. Rellergert, S. Sullivan, K. Chen, and E.R. Hudson, "Sympathetic Vibrational Cooling of Translationally Cold Molecules", P45, International Conference on Laser Spectroscopy, Berkeley, CA.
2. E.R. Hudson, "MOTION Trap: A Hybrid Neutral-Ion Environment," Fundamental Science and Applications of Ultracold Polar Molecules, KITP, Santa Barbara, CA.
3. E.R. Hudson, "Sympathetic cooling of molecules with laser cooled atoms," New Science with Ultracold Polar Molecules, KITP, Santa Barbara, CA.
4. E.R. Hudson, "Sympathetic cooling of molecules with laser cooled atoms," IOTA-COST Workshop on molecular ions, Arosa, Switzerland.
5. E.R. Hudson, "Sympathetic cooling of molecules with laser cooled atoms," Cold and Ultracold Molecules Workshop, Granada, Spain.

Number of Presentations: 5.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received Paper

TOTAL:

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received Paper

TOTAL:

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

(d) Manuscripts

Received Paper

TOTAL:

Number of Manuscripts:

Books

Received Paper

TOTAL:

Patents Submitted

Patents Awarded

Awards

Eric R. Hudson, UCLA Department of Physics and Astronomy, Outstanding Teaching Award

Graduate Students

| <u>NAME</u> | <u>PERCENT SUPPORTED</u> | Discipline |
|------------------------|--------------------------|------------|
| Scott T. Sullivan | 0.50 | |
| Steven J. Schowalter | 0.50 | |
| Kuang Chen | 0.00 | |
| FTE Equivalent: | 1.00 | |
| Total Number: | 3 | |

Names of Post Doctorates

| <u>NAME</u> | <u>PERCENT SUPPORTED</u> |
|------------------------|--------------------------|
| Wade G. Rellergert | 0.00 |
| Christian Schneider | 1.00 |
| K. Chen | 0.50 |
| FTE Equivalent: | 1.50 |
| Total Number: | 3 |

Names of Faculty Supported

| <u>NAME</u> | <u>PERCENT SUPPORTED</u> | National Academy Member |
|------------------------|--------------------------|-------------------------|
| Eric R. Hudson | 0.08 | |
| FTE Equivalent: | 0.08 | |
| Total Number: | 1 | |

Names of Under Graduate students supported

| <u>NAME</u> | <u>PERCENT SUPPORTED</u> | Discipline |
|------------------------|--------------------------|------------|
| Prateek Puri | 1.00 | Physics |
| Seth Linker | 0.00 | Physics |
| Krish Bhutwala | 1.00 | Physics |
| FTE Equivalent: | 2.00 | |
| Total Number: | 3 | |

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: 2.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:..... 2.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:..... 2.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):..... 2.00

Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:..... 0.00

The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense 0.00

The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields:..... 1.00

Names of Personnel receiving masters degrees

NAME

Steven J. Schowalter

Total Number:

1

Names of personnel receiving PHDs

NAME

Scott T. Sullivan

Kuang Chen

Total Number:

2

Names of other research staff

NAME

PERCENT SUPPORTED

FTE Equivalent:

Total Number:

Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

See Attachment

Technology Transfer

Foreword and Statement of the problem studied

The primary focus of our effort was to produce ultracold molecular ions via sympathetic cooling with laser cooled atoms. Building on the atom-ion chemistry and molecular spectroscopy work of previous years, we performed several proof-of-principle experiments that have demonstrated, for the first time, sympathetic cooling of both the translational and vibrational motion of BaCl^+ molecules through collisions with ultracold gases. These experimental results have been indicated that the proposed cooling method is as efficient as expected. We have also spent significant time understanding the details of the cooling mechanisms so that the experimental system can be optimized. Work has now turned to the next generation of experiments.

In what follows we give brief description of each effort undertaken during this project year.

Summary of the most important results

Introduction to the experimental system (MOTION trap): The construction of the original MOTION trap system was completed in late 2010 and is shown in the accompanying photo and figure below. At the center of the vacuum chamber, denoted by the blue arrow, is ^{40}Ca magneto-optical trap (MOT) collocated with a linear quadrupole radio-frequency ion trap (LQIT) system. The ^{40}Ca MOT laser beams (blue) are evidenced by scattered light from the optics. The necessary computer and laser controls can be seen in the background. Using this system we have studied the MOTION trap environment as detailed below.

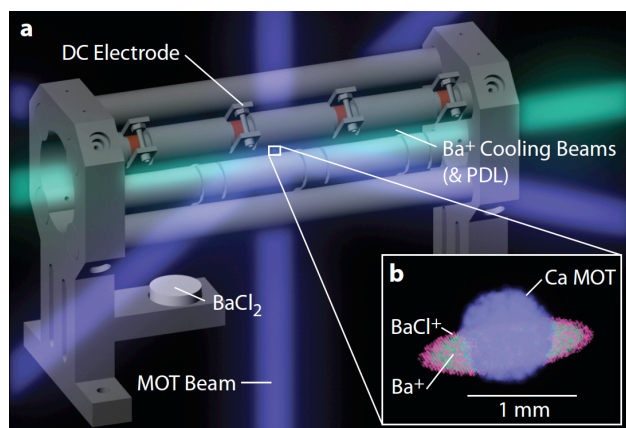
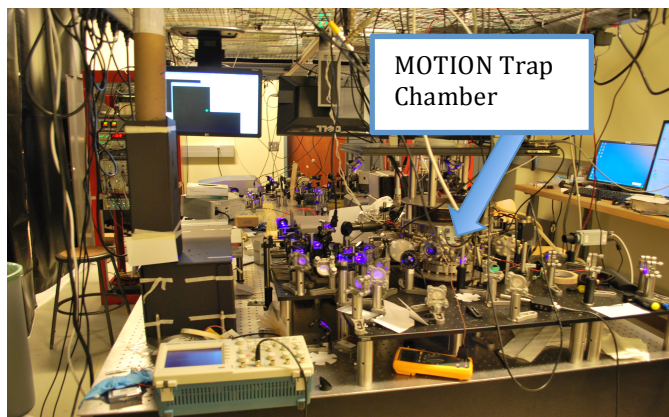


Figure 1. First-generation MOTION trap system. Molecular and/or atomic ions that have been produced by laser ablation of a solid target are trapped in the middle chamber of the linear quadrupole trap, while ultracold atoms are collected in a MOT. The ultracold atoms quickly cool the atomic/molecular ion's external and internal degrees of freedom.

Work towards understanding the kinematics of sympathetic cooling in an ion trap: Since the work of Dehmelt over forty years ago, it has been known that sympathetic cooling in an ion trap environment is much more complicated than in a static trap. However, an accurate, complete description of the effects observed in these systems has never been presented. We have explored

these effects both theoretically and experimentally and have now developed a complete understanding of the processes at play. These effects stem from the occurrence of collisions in the presence of the radio-frequency (rf) trapping voltages of the ion trap and allow for energy to be coupled from the rf trap field into the motion of the ion. This work has resulted in one manuscript to-date and another is in preparation: K. Chen et al., Phys. Rev. Lett. **110**, 173003 (2013) (Editor’s suggestion) and K. Chen et al., submitted, (2013). Finally, we believe this work will continue to be fruitful for the foreseeable future, as we have recently discovered that these effects lead to a rich system exhibiting power law energy distributions and non-equilibrium steady state physics.

Demonstration of the production of cold molecular ions: Using the MOTION trap system we implemented a two-stage sympathetic cooling procedure to cool BaCl^+ . Trapped BaCl^+ ions were immersed in laser-cooled clouds of Ba^+ ions and Ca atoms. Due to the strong Coulomb interaction, the Ba^+ ions quickly cool the molecular ion translation motion, while the neutral Ca atoms cool the BaCl^+ molecular ion internal degrees of freedom (rovibrational quantum states) via short-range atom-ion sympathetic cooling collisions. As shown in Fig. 2, which plots the results of a photodissociative thermometry measurement, the molecular ions have been cooled to the vibrational ground state with $\sim 90\%$ probability. This work demonstrates the principles of the ultimate goal of this proposal. We expect this work to be of high impact in the field as it represents a simple, generic method for producing cold molecular ions, and thus enables a host of technologies and fundamental physics, as outlined in our proposal. This work was reported in a manuscript in Nature. [W.G. Rellergert et al., “Sympathetic vibrational cooling of translationally cold molecules,” Nature **495**, 490 (2012)].

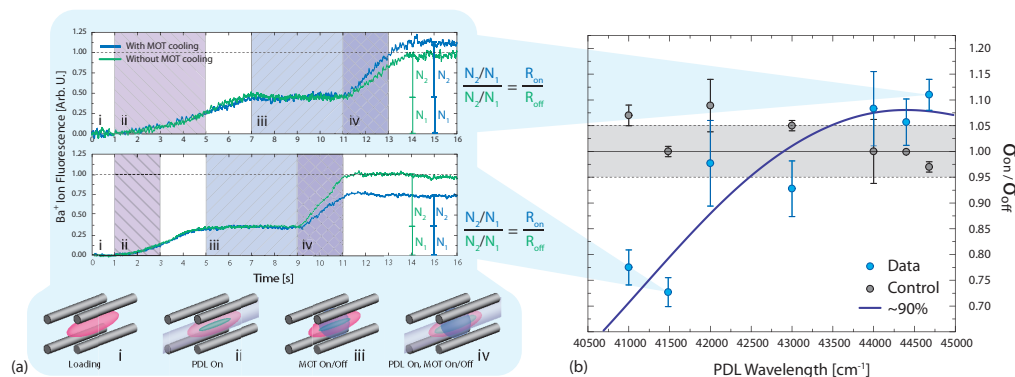


Fig. 2. (Left) Fluorescence of Ba^+ ions produced via photodissociation of BaCl^+ as a function of time with and without the MOT. The difference in final fluorescence values is a direct result of the BaCl^+ internal state temperature and can be used to calculate the molecular vibrational populations. (Right) Results of photodissociation thermometry showing $\sim 90\%$ ground state occupation.

Construction of the next generation MOTION trap: Using what was learned in the sympathetic cooling experiments, we have redesigned and constructed the second generation of the MOTION trap. This system will afford single ion imaging and colder final temperatures for the molecular ions. We have also added time-of-flight mass spectrometer to the system, which will provide a paradigm shift in how we study of ultracold atom-ion chemistry. This work was a significant undertaking and has required the development of novel rf electronics, all of which will be documented in a technical paper.

This system became fully-operational in November of 2013. Already, we it has produced exciting results, demonstrating both single ion imaging resolution and new route to improving mass spectrometry. We are in the process of applying for patents for the improved mass spectrometry technique. We expect this system to enable a new level of control in both the production of ultracold molecular ions and cold aotm-ion chemistry.

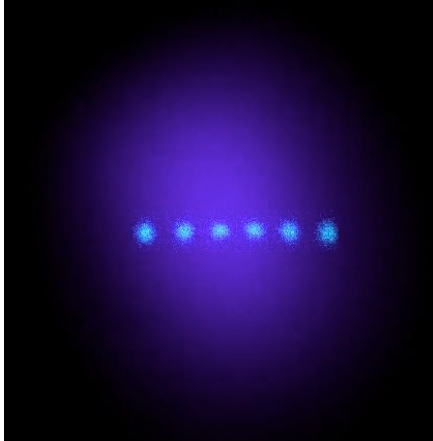


Figure 3. A string of trapped Yb-ions immersed in a Ca MOT in the second-generation of the MOTION trap.

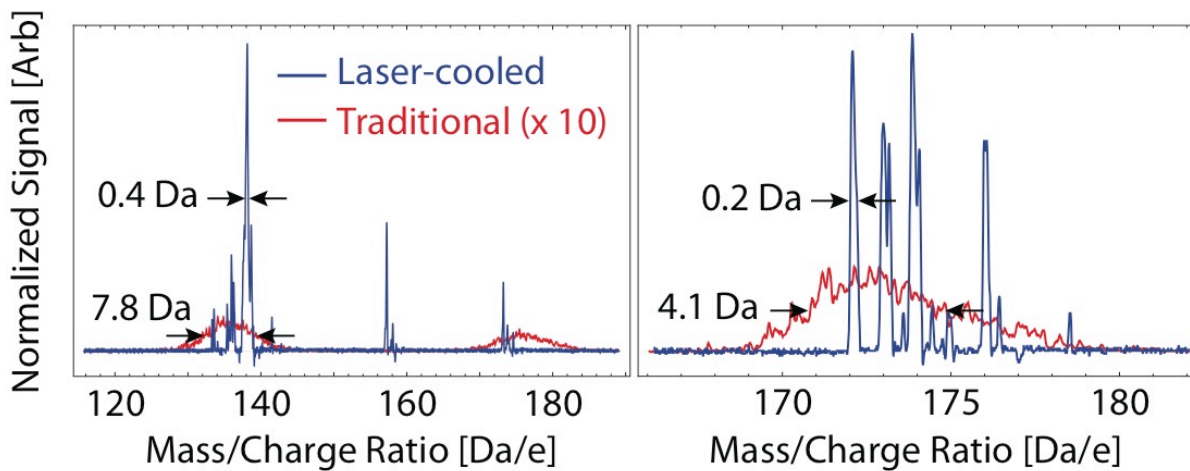


Figure 4. Traditional vs. optimized laser-cooled mass spectra. Left: mass spectrum of the products of laser ablating solid BaCl_2 while laser-cooling $^{138}\text{Ba}^+$. Right: mass spectrum of the products of laser ablating Yb metal while laser-cooling $^{172}\text{Yb}^+$. Both the mass resolution and detection efficiency are improved by over an order of magnitude. (We are in the process of applying for a patent for this technique. We ask that the reader keep this communication in confidence.)

Human resource development: This project has trained two postdocs, four graduate students, and seven undergraduate students. The postdoc and graduate students have become experts in modern AMO techniques. One of the postdocs has accepted a permanent position in the quantum technologies division of the Jet Propulsion Laboratory. Two of the graduate students defended their PhD's in September 2013 and the rest have advanced to PhD candidacy. Six of the

undergraduates trained on this project are from underrepresented backgrounds; four have gone on to graduate school at top universities and the other two plan to upon graduation.