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Duke University
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a. REPORT	b. ABSTRACT	c. THIS PAGE			Kenneth R. Brown
UU	UU	UU	UU		19b. TELEPHONE NUMBER 919-660-1376

RPPR Final Report
as of 15-Oct-2023

Agency Code: 21XD

Proposal Number: 73260PE

Agreement Number: W911NF-18-1-0258

INVESTIGATOR(S):

Name: Kenneth R. Brown
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Final Report for Period Beginning 05-Jul-2018 and Ending 28-May-2021

Title: Control and Spectroscopy of Single Molecular Ions

Begin Performance Period: 05-Jul-2018

End Performance Period: 28-May-2021

Report Term: 0-Other

Submitted By: Kenneth R. Brown

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Distribution Statement: 1-Approved for public release; distribution is unlimited.

STEM Degrees: 0

STEM Participants:

Major Goals: The goals of this project were the continued development of spectroscopy techniques for molecular ions co-trapped with atomic ions. This work focused on methods for molecular ion spectroscopy and the preparation of known molecular states.

Accomplishments: Key accomplishments included a perspective article that laid out current and future spectroscopy techniques. Some of these techniques have now been implemented by other groups including David Patterson at UC Santa Barbara.

A key direction was the testing of the sympathetic cooling of the internal states of molecular ions by interaction with laser-cooled neutral atoms. We were able to show that the interaction of the cooled K atoms and Ca+ ions was negligible. The charge exchange observed is primarily driven by photoexcitation.

In collaboration with researchers at UCLA and Emory, we examined the prospects of dipole photon quantum logic using Ca+ and CaO+ and trapped Ca+ and CaO+ together to start the exploration of this work experimentally.

Training Opportunities: The work trained two postdocs and one graduate student.

Results Dissemination: The work was disseminated by publications and project presentations.

Notable presentations include:
Ion Atom Hybrid System: K and Ca+, Few-body and Collective Many-body Behavior with Charge Impurities in Atomic Quantum Gases Workshop, San Felieu Guixol, Spain, July 19, 2018

Towards sympathetic translational and rotational cooling of CaH+ in an ion-atom hybrid trap, Jyothi Saraladevi, North American Conference on Trapped Ions, University of Maryland, July 22-26, 2019

Honors and Awards: Kenneth Brown was named a Fellow of the American Physical Society.

Protocol Activity Status:

Technology Transfer: Nothing to Report

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PARTICIPANTS:

Participant Type: PD/PI
Participant: Kenneth Brown Brown
Person Months Worked: 3.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: Eric Pretzsch
Person Months Worked: 6.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
Participant: Jyothi Saraladevi
Person Months Worked: 15.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
Participant: Lu Qi
Person Months Worked: 11.00 **Funding Support:**
Project Contribution:
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ARTICLES:

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Journal: The Journal of Physical Chemistry Letters
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Volume: 9 Issue: 19 First Page #: 5797
Date Submitted: 5/31/19 12:00AM Date Published: 9/1/18 12:00AM
Publication Location:
Article Title: Spectroscopy of Molecular Ions in Coulomb Crystals
Authors: Aaron T. Calvin, Kenneth R. Brown
Keywords: cold molecular ions, molecular ion spectroscopy, laser cooling
Abstract: We examine the use of laser-cooled atomic ions and sympathetically cooled molecular ions in Coulomb crystals for molecular spectroscopy. Coulomb crystals are well-isolated environments that provide localization and long storage times for sensitive measurements of weak signals and cold temperatures for precise spectroscopy. Coulomb crystals of molecular and atomic ions enable the detection of single-photon molecular ion transitions at a range of wavelengths by a change in atomic ion fluorescence at visible wavelengths. We give an overview of the state of the art from action spectroscopy to quantum logic spectroscopy for a wide range of molecular transitions from rotational sublevels separated by 10–7 cm–1 to rovibronic transitions at 25?000 cm–1. We emphasize how this system allows for unparalleled control of the molecular ion state for precision spectroscopy with applications in astrochemistry and fundamental physics. We conclude with an outlook of the use of this control in reactions.
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Volume: 90 Issue: 10 First Page #: 103201
Date Submitted: 8/29/20 12:00AM Date Published: 10/1/19 4:00AM
Publication Location:
Article Title: A hybrid ion-atom trap with integrated high resolution mass spectrometer
Authors: S. Jyothi, Kisra N. Egodapitiya, Brad Bondurant, Zhubing Jia, Eric Pretzsch, Piero Chiappina, Gang Shu
Keywords: charge exchange, trapped ions, photochemistry, mangeto-optical trap, laser cooling, laser-cooled mass spectrometry
Abstract: In this article, we describe the design, construction, and implementation of our ion-atom hybrid system incorporating a high resolution time of flight mass spectrometer (TOFMS). Potassium atoms (39K) in a magneto optical trap and laser cooled calcium ions (40Ca+) in a linear Paul trap are spatially overlapped, and the combined trap is integrated with a TOFMS for radial extraction and detection of reaction products. We also present some experimental results showing interactions between 39K+ and 39K, 40Ca+ and 39K+, as well as 40Ca+ and 39K pairs. Finally, we discuss prospects for cooling CaH+ molecular ions in the hybrid ion-atom system.
Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info
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Volume: 22 Issue: 19 First Page #: 10870
Date Submitted: 8/29/20 12:00AM Date Published: 4/22/20 4:00AM
Publication Location:
Article Title: Photon-mediated charge exchange reactions between
Authors: Hui Li, S. Jyothi, Ming Li, Jacek Klos, Alexander Petrov, Kenneth R Brown, Svetlana Kotochigova
Keywords: ion traps, magneto-optical trap, laser cooling, charge exchange, photochemistry
Abstract: We present experimental evidence of charge exchange between laser-cooled potassium 39K atoms and calcium 40Ca+ ions in a hybrid atom–ion trap and give quantitative theoretical explanations for the observations. The 39K atoms and 40Ca+ ions are held in a magneto-optical (MOT) and a linear Paul trap, respectively. Fluorescence detection and high resolution time of flight mass spectra for both species are used to determine the remaining number of 40Ca+ ions, the increasing number of 39K+ ions, and 39K number density as functions of time. Simultaneous trap operation is guaranteed by alternating periods of MOT and 40Ca+ cooling lights, thus avoiding direct ionization of 39K by the 40Ca+ cooling light. We show that the K–Ca+ charge-exchange rate coefficient increases linearly from zero with 39K number density and the fraction of 40Ca+ ions in the 4p 2P1/2 electronically-excited state. Combined with our theoretical analysis, we conclude that these data can only be explained by a process that
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Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Chemistry Chemical Physics
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Volume: 22 Issue: 43 First Page #: 24964
Date Submitted: 9/28/23 12:00AM Date Published:
Publication Location:
Article Title: Dipole–phonon quantum logic with alkaline-earth monoxide and monosulfide cations
Authors: Michael Mills, Hao Wu, Evan C. Reed, Lu Qi, Kenneth R. Brown, Christian Schneider, Michael C. Heave
Keywords: cold molecular ions, molecular qubits, dipole photon quantum logic
Abstract: Dipole–phonon quantum logic (DPQL) leverages the interaction between polar molecular ions and the motional modes of a trapped-ion Coulomb crystal to provide a potentially scalable route to quantum information science. Here, we study a class of candidate molecular ions for DPQL, the cationic alkaline-earth monoxides and monosulfides, which possess suitable structure for DPQL and can be produced in existing atomic ion experiments with little additional complexity. We present calculations of DPQL operations for one of these molecules, CaO+, and discuss progress towards experimental realization. We also further develop the theory of DPQL to include state preparation and measurement and entanglement of multiple molecular ions.
Distribution Statement: 1-Approved for public release; distribution is unlimited.
Acknowledged Federal Support: **Y**

Partners

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I certify that the information in the report is complete and accurate:
Signature: Kenneth Ray Brown
Signature Date: 9/29/23 12:47AM

Control and Spectroscopy of Single Molecular Ions
W911NF1810258

Abstract: The combination of atomic ions and cold molecular ions offers an opportunity for high-precision spectroscopy and molecular ion control. The work had two technical focuses: 1) the development of an apparatus that combined molecular ions, laser-cooled atomic ions, and laser-cooled atoms; and 2) the construction of a system for quantum logic between atomic and molecular ions. We were able to demonstrate that for the first system that molecular ions, laser-cooled ions, and laser-cooled atoms could be combined. For the quantum logic system, we prepared the necessary control lasers and demonstrated the co-trapping of CaO+ and Ca+.

Objective 1: Exploration of molecular ion spectroscopies using cold atomic ions and molecular ions

We published a perspective paper in J. Phys. Chem. Lett. on molecular ion spectroscopies with atomic ions. We found that there are many ways to combine action spectroscopies from physical chemistry to Coulomb crystals containing atomic and molecular ions. The first author, Aaron Calvin, sponsored under the previous grant W911NF-17-1-0071, pursued these ideas during his postdoc at the UC Santa Barbara.

Objective 2: State preparation of rotational states of molecular ions by sympathetic cooling with laser-cooled neutral atoms.

We did not achieve this objective but set the ground work. The experiment is the continuation of W911NF-17-1-0071 when the PI Brown moved from the Georgia Institute of Technology to Duke University. The experiment enables the simultaneous trapping of K in a magneto-optical trap and Ca^+ and CaH^+ in a linear Paul trap. The plans is for Ca^+ to cool the motion of CaH^+ and the K cools the internal states

Year 1 – The apparatus was moved from Georgia Tech to Duke and refurbished.
Year 2 – Data was collected for the interaction between the atomic ion and neutral atoms. The apparatus was described in a Review of Scientific Instruments paper. We worked with collaborators at Temple University to understand the charge exchanged between K and Ca+ ions. The results were published in Phys. Chem. Chem. Phys. The reaction is energetically favorable in the ground stated but kinetically slow. The measured charge exchange occurs through an excited stated and is sufficiently slow that we can continue towards molecular cooling.
Finding: Trapped Ca+ and K MOT can co-exist by stroboscopically applying laser beams.
Finding: Ca+ - K charge exchange is photon mediated and occurs at a time scale of tens of second for typical MOT beams

Year 3 – We examined the inclusion of molecular ions into the trap.
Finding: CaH^+ can be sympathetically cooled by Ca^+ in the presence of a low-density K MOT.
Finding: Laser-cooled ion trap ejection into a short time-of-flight mass spectrometer is sufficient to distinguish K^+, Ca^+, and CaH^+

Objective 3: Quantum logic between an atomic ion and a molecular ion.

Year 1-2 Objective 3 took a back seat to Objective 2 after the move from Georgia Tech. The experiment had an arm of the vaccum chamber for the creation of BH^+ by ablation of B in a jet of H_2. After the move the ablation target was found to be out of position. We worked to reset up sideband cooling of Ca^+ and the associated lasers.

Year 3 UCLA researchers reached out to us about the possibility of dipole-phonon quantum logic spectroscopy with CaO+ and Ca+. This allows for the transfer of a single molecular dipole onto the motion of an atomic ion. We developed theory to examine the optimal adiabatic sweep for transferring the excitation given known trap imperfections. We also co-trapped Ca+ and CaO+ and began optimizing the sympathetic sideband cooling.
Finding – The lambda doublet splitting of CaO+ is predicted to be near 450 kHz within the range of secular frequencies obtainable in ion traps.
Finding – Modest heating rates observed in the ion trap are compatible with the adiabatic transfer of the dipole excitation to the atomic motion.