REPORT DOCUMENTATION PAGE

Form Approved OMB NO. 0704-0188

1. REPORT DATE (DD-MM-YYYY)	2. REPORT TYPE	3. DATES COVERED (From - To)
05-05-2023	Final Report	18-Mar-2019 - 30-Sep-2022

4. TITLE AND SUBTITLE
Final Report: Quantum control and spectroscopy of a polyatomic molecular ion

5a. CONTRACT NUMBER
W911NF-19-1-0172

5b. GRANT NUMBER

5c. PROGRAM ELEMENT NUMBER
611102

6. AUTHORS

5d. PROJECT NUMBER

5e. TASK NUMBER

5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAMES AND ADDRESSES
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8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS (ES)
U.S. Army Research Office
P.O. Box 12211
Research Triangle Park, NC 27709-2211

10. SPONSOR/MONITOR'S ACRONYM(S)
ARO

11. SPONSOR/MONITOR'S REPORT NUMBER(S)
74703-PE.3

12. DISTRIBUTION AVAILIBILITY STATEMENT
Approved for public release; distribution is unlimited.

13. SUPPLEMENTARY NOTES
The views, opinions and/or findings contained in this report are those of the author(s) and should not contrued as an official Department of the Army position, policy or decision, unless so designated by other documentation.

14. ABSTRACT

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	15. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			David Leibrandt
UU	UU	UU	UU		19b. TELEPHONE NUMBER: 303-497-7292

RPPR Final Report
as of 12-May-2023

Agency Code: 21XD

Proposal Number: 74703PE **Agreement Number: W911NF-19-1-0172**
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Report Date: 31-Dec-2022 Date Received: 05-May-2023
Final Report for Period Beginning 18-Mar-2019 and Ending 30-Sep-2022
Title: Quantum control and spectroscopy of a polyatomic molecular ion
Begin Performance Period: 18-Mar-2019 **End Performance Period:** 30-Sep-2022
Report Term: 0-Other

Submitted By: Ph.D. Chin-wen Chou Email: chin-wen.chou@nist.gov
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STEM Degrees: **STEM Participants:**

Major Goals: 1. Demonstrating quantum entanglement between an atomic ion and a molecular ion. Designing and constructing a cryogenic ion trapping apparatus,
2. Increasing the experimental data rate with deterministic pure state preparation of CaH+ in the room-temperature apparatus,
3. Trapping Ca+ in a cryogenic ion trap and characterizing the trap performance and cryocooler-induced vibrations. Applying the techniques to a polyatomic molecular ion N2H+.

Accomplishments: Year 1: Demonstrating quantum entanglement between an atomic ion and a molecular ion. We have demonstrated entanglement between an atomic ion qubit and a molecular ion qubit via Cirac-Zoller type interaction. Specifically, starting with both the Ca+ and CaH+ ions prepared in pure quantum states and one of their shared harmonic motional modes cooled near the ground state, a pi/2 pulse on the motion-exciting sideband of a molecular qubit transition prepares the CaH+ and the motional mode in an entangled state. The motional state is then mapped to the atomic qubit with a pi pulse on the motion-reducing sideband of the atomic qubit transition, thus entangling the Ca+ and the CaH+ ions. The atomic qubit consists of one level in the 4s S1/2 state and one in the 3d D5/2 state of Ca+, connected by a transition at ~411 THz. We had chosen two distinct sets of molecular levels as the molecular qubits. One set is within the J = 2 rotational manifold and connected by a transition with ~13 kHz frequency. The levels in the other set are in the J = 2 and J = 0 rotational manifolds, linked by a ~855 GHz transition. We aim to demonstrate the broad selection of transition frequencies offered by molecules.
The entanglement is verified by measuring the population in the target basis states and the contrast of the parity scan where the phases of atomic and molecular analysis π/2 pulses are varied. The fidelity between the produced states with the target Bell states can then be derived. For the case when both the qubit states are in the J = 2 rotational manifold, we attain a fidelity of 0.87(3). When the qubit states are in J = 2 and J = 0 manifolds, the fidelity can reach 0.76(4). In both cases, the fidelities indicate entanglement between the atom and the molecule. When

investigating the sources of infidelity, we find that the state-preparation-and-measurement (SPAM) is the dominating source. The lower fidelity when the molecular qubit states are in two different rotational manifolds is mainly attributed to the extra pulses required and imperfect coherence in the frequency comb pulses.
Year 2: Increasing the experimental data rate with deterministic pure state preparation of CaH+ in the room-temperature apparatus.
Toward the goal of deterministic pure state preparation of CaH+, the mechanisms that could affect the molecular states need to be well understood. In our work on precision rotational spectroscopy of CaH+ [chou2020], the trap rf electric field had been identified as the leading source of uncertainty. We performed systematic characterization of the trap rf electric field and developed an analytical model that describes the effect of such fields on the molecular levels. That allowed us to understand and control the effects of the fields. Equipped with better understanding of the effect of trap RF electric fields on the molecular levels and transitions, we have demonstrated Hertz-level resolution rotational spectroscopy and efficient population transfer between rotational manifolds of CaH+ by improving the coherence of our frequency comb. We have also incorporated a 285-GHz microwave source in the experimental setup and successfully driven the J = 0 to J = 1 transition in CaH+. The frequency comb and the 285-GHz source enable reliable coherent transfer of molecular population between rotational manifolds with even and odd principal quantum number, which is essential for deterministic pure state preparation of CaH+.
With the understanding of the effects of the rf electric field on the molecular levels, we decided to increase the bias magnetic field from 3.6 Gauss to 6.5 Gauss, which lifted degeneracy in some molecular levels and improved the efficiency of quantum-logic pumping. In place of a frequency comb, which requires significant upkeep, we have introduced a 570 GHz microwave source to move the population between J = 1 and J = 2 manifolds. Along with the 285-GHz microwave source, the molecular population can be controllably moved within the subspace consisting of J = 0, 1, and 2 manifolds. We have combined improved pumping within J = 1 and J = 2 rotational manifolds and the microwave sources to develop an experimental protocol track and trap the molecular state in the rotational manifolds with J smaller or equal to 2. After the molecular state is projectively prepared in J = 1, after some operations for control and/or spectroscopy purposes, the molecular state in J = 1 is checked with nondestructive detection. A positive detection triggers operations moving the molecular population into the desired state and the experimental sequence carries on. If the detection results indicate that the molecular population is not in J = 1 anymore, the control system will search for it in J = 2 sublevels and then in the J = 0 manifold. If the population is found in J = 0 or J = 2 manifolds, the 285 GHz or 570 GHz microwave source, respectively, will be used to move it back to the desired J = 1 sublevel, after which the experimental sequence can resume. This procedure repeats until the population cannot be found in the J =0, 1 or 2 manifolds, at which point the control system switches to perform projective state preparation, with the state preparation success rate determined by the approximately blackbody radiation from the trap environment of the molecule.
The above sequence enabled significant improvement, from few percent to >50 percent, of the average fraction of time over which the experimental control system keeps track of the molecular population and experimental sequences can be performed. The increased data rate will greatly accelerate subsequent investigations.
Year 1: designing and constructing a cryogenic ion trapping apparatus; Year 3: Trapping Ca+ in a cryogenic ion trap and characterizing the trap performance and cryocooler-induced vibrations. Applying the techniques to a polyatomic molecular ion N2H+.
Toward studying Ca+ in the cryogenic ion trap, we have finished assembling the apparatus, tested operation of the trap at room temperature, and loaded Ca+ and various molecular species including ones with mass-to-charge ratio 28 (likely N2+) and 29 (likely N2H+). Testing and improvements of the molecular beam source took some time and the optimization for loading certain molecular species is still underway. For the ionization, we have tested pulsed lasers with wavelengths ranging from UV to near infrared, pulse energies from hundreds of microjoules to few millijoules, and pulse durations from hundreds of picoseconds to a few nanoseconds. The ion production using pulsed lasers was not efficient. Using loading success rate in a trap as the signal for ionization efficiency was not successful. With the guidance from Prof. Lewandowski at JILA, we introduced a multichannel plate (MCP) to detect ions produced by an electron gun. Such a setup allowed us to produce molecular ions and tune up the molecular flux from the molecular beam source. We are in the process of adding an ion detection chamber to the trap apparatus. The plan is to ionize neutral molecules from the beam source with an electron gun embedded in one of the endcaps on the trap. We are currently modifying the trap apparatus to accommodate cryogenic operations and the detection chamber for the molecular beam. After the system is tuned up for Ca+ and CaH+, we will move on to studying N2H+.

Training Opportunities: Two graduate student, Dalton Chaffee and Zhimin Liu, and three postdoctoral researchers, Alejandra Collopy, Julian Schmidt, and Yu Liu worked on the project and received training on quantum state control of single trapped molecular ions.

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Results Dissemination: Results of the project are presented in conferences and published to peer-reviewed journals as listed below.
Selected conference presentations:
1. Gordon conference on Atomic Physics, Newport, RI, 2019.
2. New frontiers in cold molecules, Cambridge, MA, 2019
3. The 50th Winter Colloquium on the Physics of Quantum Electronics, Snowbird, UT, 2020
4. Seminar on Precision Physics and Fundamental Symmetries, virtual, 2020
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Publications:
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[lin2020] Y. Lin, D. R. Leibrandt, D. Leibfried, and C. W. Chou, Quantum entanglement between an atom and a molecule, Nature 581, 273 (2020).
[chou2020] C. W. Chou, A. L. Collopy, C. Kurz, Y. Lin, M. E. Harding, P. N. Plessow, T. Fortier, S. Diddams, D. Leibfried, and D. R. Leibrandt, Frequency-comb spectroscopy on pure quantum states of a single molecular ion, Science 367, 1458 (2020).

Honors and Awards: 2021 NIST BRONZE MEDAL AWARD

Protocol Activity Status:

Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
Participant: Julian Schmidt
Person Months Worked: 6.00 **Funding Support:**
Project Contribution:
National Academy Member: N

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

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
Participant: Yu Liu
Person Months Worked: 6.00 **Funding Support:**
Project Contribution:
National Academy Member: N

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ARTICLES:

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Science
Publication Identifier Type: DOI Publication Identifier: 10.1126/science.aba3628
Volume: 367 Issue: 6485 First Page #: 1458
Date Submitted: 8/27/20 12:00AM Date Published: 3/1/20 7:00AM
Publication Location:
Article Title: Frequency-comb spectroscopy on pure quantum states of a single molecular ion
Authors: C. W. Chou, A. L. Collopy, C. Kurz, Y. Lin, M. E. Harding, P. N. Plessow, T. Fortier, S. Diddams, D. Leib
Keywords: molecular spectroscopy, quantum control, ions
Abstract: Spectroscopy is a powerful tool for studying molecules and is commonly performed on large thermal molecular ensembles that are perturbed by motional shifts and interactions with the environment and one another, resulting in convoluted spectra and limited resolution. Here, we use quantum-logic techniques to prepare a trapped molecular ion in a single quantum state, drive terahertz rotational transitions with an optical frequency comb, and read out the final state nondestructively, leaving the molecule ready for further manipulation. We can resolve rotational transitions to 11 significant digits and derive the rotational constant of 40CaH+ to be BR = 142 501 777.9(1.7) kilohertz. Our approach is suited for a wide range of molecular ions, including polyatomics and species relevant for tests of fundamental physics, chemistry, and astrophysics.
Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info
Acknowledged Federal Support: **Y**

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Nature
Publication Identifier Type: DOI Publication Identifier: 10.1038/s41586-020-2257-1
Volume: 581 Issue: 7808 First Page #: 273
Date Submitted: 8/27/20 12:00AM Date Published: 5/1/20 6:00AM
Publication Location:
Article Title: Quantum entanglement between an atom and a molecule
Authors: Yiheng Lin, David R. Leibrandt, Dietrich Leibfried, Chin-wen Chou
Keywords: molecules, ions, entanglement, quantum transduction
Abstract: Conventional information processors convert information between different physical carriers for processing, storage and transmission. It seems plausible that quantum information will also be held by different physical carriers in applications such as tests of fundamental physics, quantum enhanced sensors and quantum information processing. Quantum controlled molecules, in particular, could transduce quantum information across a wide range of quantum bit (qubit) frequencies—from a few kilohertz for transitions within the same rotational manifold1, a few gigahertz for hyperfine transitions, a few terahertz for rotational transitions, to hundreds of terahertz for fundamental and overtone vibrational and electronic transitions—possibly all within the same molecule. Here we demonstrate entanglement between the rotational states of a 40CaH+ molecular ion and the internal states of a 40Ca+ atomic ion2. We extend methods used in quantum logic spectroscopy1,3 for pure-state initialization, las
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Partners

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I certify that the information in the report is complete and accurate:
Signature: Chin-wen Chou
Signature Date: 5/5/23 6:23PM

Major goals

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Accomplished under Goals

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