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12. DISTRIBUTION AVAILIBILITY STATEMENT

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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: a. REPORT	b. ABSTRACT	c. THIS PAGE	17. LIMITATION OF ABSTRACT	15. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
UU	UU	UU	UU		Wolfgang Ketterle
					19b. TELEPHONE NUMBER 617-253-6815

RPPR Final Report
as of 16-Mar-2023

Agency Code: 21XD

Proposal Number: 63535PERIP
Agreement Number: W911NF-13-1-0411

INVESTIGATOR(S):

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DUNS Number: 001425594
EIN: 042103594
Report Date: 25-Feb-2015
Date Received: 13-Mar-2023
Final Report for Period Beginning 26-Aug-2013 and Ending 25-Nov-2014
Title: A Laser System for Spin-Dependent Optical Lattices and Polar Molecules
Begin Performance Period: 26-Aug-2013
End Performance Period: 25-Nov-2014
Report Term: 0-Other
Submitted By: Timothy Swager
Email: tswager@mit.edu
Phone: (617) 253-4423
Distribution Statement: 1-Approved for public release; distribution is unlimited.

STEM Degrees: 0
STEM Participants:

Major Goals: The equipment purchase supported our program in two labs: The study of ultracold NaLi molecules, and investigations of Rb atoms in optical lattices.

Accomplishments: This grant allowed us to buy a tunable Titanium-Sapphire laser which could be used in two different laboratories for two different purposes.

In our ultracold molecule lab, we used the tunable laser first to perform spectroscopy of the triplet state of NaLi. This work identified a pathway to crate the triplet ground state via two-photon photoassociation. Using triplet ground state NaLi, we have made a number of breakthrough observations, including the first sympathetic cooling of an ultracold molecule, and the control of chemical reactions via magnetic fields (see upload) ,

In our ultracold rubidium lab, we used the tunable laser for spin-dependent lattices. This allowed us to realize a new quantum state of matter, a Mott insulator of repulsively bound pairs. We had obtained preliminary results already many years earlier, but then decided to work on different projects first. The 2022 PRL is the realization of what we had proposed in the DURIP proposal (see upload).

Training Opportunities: The equipment has benefitted the research and education of several graduate students. Experiments in atomic physics provide training for students and prepares them for a career in science and technology.

Results Dissemination: The main mechanism for dissemination is through publications and talks at conferences.

Honors and Awards: Nothing to Report

Protocol Activity Status:

Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: PD/PI
Participant: Wolfgang Ketterle

RPPR Final Report
as of 16-Mar-2023

Person Months Worked: 1.00 **Funding Support:**
Project Contribution:
National Academy Member: Y

ARTICLES:

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Chemistry Chemical Physics
Publication Identifier Type: DOI Publication Identifier: 10.1039/C7CP08481A
Volume: 20 Issue: 7 First Page #: 4739
Date Submitted: 3/13/23 12:00AM Date Published:
Publication Location:
Article Title: Two-photon spectroscopy of the NaLi triplet ground state
Authors: Timur M. Rvachov, Hyungmok Son, Juliana J. Park, Sepehr Ebadi, Martin W. Zwierlein, Wolfgang Kette
Keywords: NaLi, spectroscopy
Abstract: We employ two-photon spectroscopy to study the vibrational states of the triplet ground state potential (a3S+) of the 23Na6Li molecule. Pairs of Na and Li atoms in an ultracold mixture are photoassociated into an excited triplet molecular state, which in turn is coupled to vibrational states of the triplet ground potential. Vibrational state binding energies, line strengths, and potential fitting parameters for the triplet ground a3S+ potential are reported. We also observe rotational splitting in the lowest vibrational state.
Distribution Statement: 1-Approved for public release; distribution is unlimited.
Acknowledged Federal Support: **Y**

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Review Letters
Publication Identifier Type: DOI Publication Identifier: 10.1103/PhysRevLett.119.143001
Volume: 119 Issue: 14 First Page #:
Date Submitted: 3/13/23 12:00AM Date Published: 10/1/17 4:00AM
Publication Location:
Article Title: Long-Lived Ultracold Molecules with Electric and Magnetic Dipole Moments
Authors: Timur M. Rvachov, Hyungmok Son, Ariel T. Sommer, Sepehr Ebadi, Juliana J. Park, Martin W. Zwierleir
Keywords: Polar molecules, electric dipole moment, ultracold molecules
Abstract: We create fermionic dipolar 23Na6Li molecules in their triplet ground state from an ultracold mixture of 23Na and 6Li. Using magnetoassociation across a narrow Feshbach resonance followed by a two-photon stimulated Raman adiabatic passage to the triplet ground state, we produce 3 × 104 ground state molecules in a spin-polarized state. We observe a lifetime of 4.6 s in an isolated molecular sample, approaching the p-wave universal rate limit. Electron spin resonance spectroscopy of the triplet state was used to determine the hyperfine structure of this previously unobserved molecular state.
Distribution Statement: 1-Approved for public release; distribution is unlimited.
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/s41586-020-2141-z
Volume: 580 Issue: 7802 First Page #: 197
Date Submitted: 3/13/23 12:00AM Date Published: 4/1/20 4:00AM
Publication Location:

Article Title: Collisional cooling of ultracold molecules

Authors: Hyungmok Son, Juliana J. Park, Wolfgang Ketterle, Alan O. Jamison

Keywords: sympathetic cooling, ultracold molecules

Abstract: Since the original work on Bose–Einstein condensation1,2, the use of quantum degenerate gases of atoms has enabled the quantum emulation of important systems in condensed matter and nuclear physics, as well as the study of many-body states that have no analogue in other fields of physics 3. Ultracold molecules in the micro- and nanokelvin regimes are expected to bring powerful capabilities to quantum emulation4 and quantum computing5, owing to their rich internal degrees of freedom compared to atoms, and to facilitate precision measurement and the study of quantum chemistry6. Quantum gases of ultracold atoms can be created using collision-based cooling schemes such as evaporative cooling, but thermalization and collisional cooling have not yet been realized for ultracold molecules. Other techniques, such as the use of supersonic jets and cryogenic buffer gases, have reached temperatures limited to above 10 millikelvin 7,8. Here we show cooling of NaLi molecules to micro- and nano

Distribution Statement: 1-Approved for public release; distribution is unlimited.
Acknowledged Federal Support: **Y**

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Science
Publication Identifier Type: DOI Publication Identifier: 10.1126/science.abl7257
Volume: 375 Issue: 6584 First Page #: 1006
Date Submitted: 3/13/23 12:00AM Date Published: 3/1/22 5:00AM
Publication Location:

Article Title: Control of reactive collisions by quantum interference

Authors: Hyungmok Son, Juliana J. Park, Yu-Kun Lu, Alan O. Jamison, Tijs Karman, Wolfgang Ketterle

Keywords: control of reactions, ultracold molecules, coherent quantum chemistry

Abstract: In this study, we achieved magnetic control of reactive scattering in an ultracold mixture of 23Na atoms and 23Na 6Li molecules. In most molecular collisions, particles react or are lost near short range with unity probability, leading to the so-called universal rate. By contrast, the Na + NaLi system was shown to have only ~4% loss probability in a fully spin-polarized state. By controlling the phase of the scattering wave function via a Feshbach resonance, we modified the loss rate by more than a factor of 100, from far below to far above the universal limit. The results are explained in analogy with an optical Fabry-Perot resonator by interference of reflections at short and long range. Our work demonstrates quantum control of chemistry by magnetic fields with the full dynamic range predicted by our models.

Distribution Statement: 1-Approved for public release; distribution is unlimited.
Acknowledged Federal Support: **Y**

RPPR Final Report
as of 16-Mar-2023

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Review Letters
Publication Identifier Type: DOI Publication Identifier: 10.1103/PhysRevLett.128.093401
Volume: 128 Issue: 9 First Page #:
Date Submitted: 3/13/23 12:00AM Date Published: 2/1/22 10:00AM
Publication Location:
Article Title: Preparation of the Spin-Mott State: A Spinful Mott Insulator of Repulsively Bound Pairs
Authors: Julius de Hond, Jinggang Xiang, Woo Chang Chung, Enid Cruz-Colón, Wenlan Chen, William Cody Bur
Keywords: Mott insulator, optical lattice
Abstract: We observe and study a special ground state of bosons with two spin states in an optical lattice: the spin- Mott insulator, a state that consists of repulsively bound pairs that is insulating for both spin and charge transport. Because of the pairing gap created by the interaction anisotropy, it can be prepared with low entropy and can serve as a starting point for adiabatic state preparation. We find that the stability of the spin- Mott state depends on the pairing energy, and observe two qualitatively different decay regimes, one of which exhibits protection by the gap.
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: Wolfgang Ketterle
Signature Date: 3/13/23 1:10AM

Final Report

Grant Title: A Laser System for Spin-dependent Optical Lattices and Polar Molecules

Grant Sponsor: ARO, DURIP, contract W911NF-13-1-0411

Award Amount: $176,990

Period: 8/26/2013 to 8/25/2014

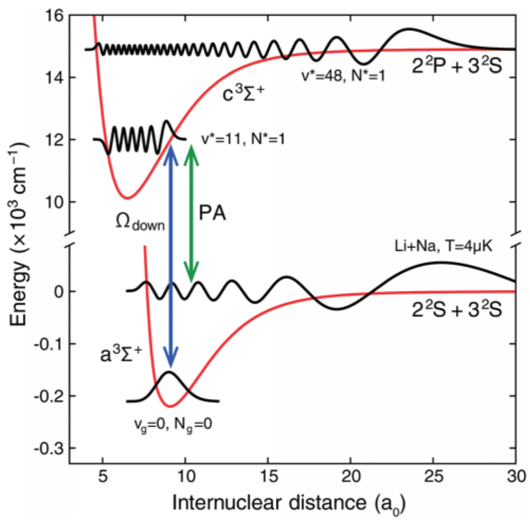
This grant allowed us to buy a tunable Titanium-Sapphire laser which could be used in two different laboratories for two different purposes.

In our ultracold molecule lab, we used the tunable laser first to perform spectroscopy of the triplet state of NaLi. This work identified a pathway to crate the triplet ground state via two-photon photoassociation. Using triplet ground state NaLi, we have made a number of breakthrough observations, including the first sympathetic cooling of an ultracold molecule, and the control of chemical reactions via magnetic fields (see below)

1. Spectroscopy of the triplet ground state of NaLi molecules

T.M. Rvachov, H. Son, J.J. Park, S. Ebadi, M.W. Zwierlein, W. Ketterle, and A.O. Jamison, *Two-Photon Spectroscopy of the $^{23}Na^{6}Li$ Triplet Ground State,* Phys. Chem. Chem. Phys. 2018, DOI:10.1039/c7cp08481a, preprint, arXiv:1712.06776, http://dx.doi.org/10.1039/C7CP08481A

We employ two-photon spectroscopy to study the vibrational states of the triplet ground state potential ($a^3\Sigma^+$) of the NaLi molecule. Pairs of Na and Li atoms in an ultracold mixture are photoassociated into an excited triplet molecular state, which in turn is coupled to vibrational states of the triplet ground potential. Vibrational state binding energies, line strengths, and potential fitting parameters for the triplet ground state potential are reported. These spectroscopic observation identified a pathway to create ultracold NaLi ground state molecules from ultracold Na and Li atoms.



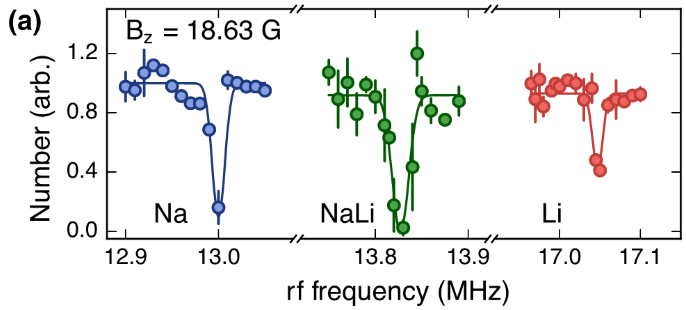
Energy potential diagram for the relevant NaLi triplet molecular states. A photoassociation laser is used to couple the initial scattering wavefunction to one of two excited states ($c^3\Sigma^+$, $v^* = 11, 48$, $N^* = 1$), which are coupled to the ground state vibrational energies with a variable frequency ‘‘downleg’’ laser used for the spectroscopic search (shown is the particular case of coupling to the ro-vibrational ground state).

2. Long-lived ultracold molecules with electric and magnetic dipole moments

T.M. Rvachov, H. Son, A.T. Sommer, S. Ebadi, J.J. Park, M.W. Zwierlein, W. Ketterle, and A.O. Jamison, Long-Lived Ultracold Molecules with Electric and Magnetic Dipole Moments, Phys. Rev. Lett. 119, 143001 (2017), https://doi.org/10.1103/PhysRevLett.119.143001

Ultracold molecules with permanent electric dipole moments have gained considerable attention in recent years as promising new systems to study quantum chemistry and quantum many-body physics. An additional magnetic dipole moment provides an extra degree of control that can be used for magnetic trapping, tuning collisions and chemical reactions, simulation of spin-lattice Hamiltonians, or, as in the case of cold magnetic atoms, direct study of magnetic dipole interaction effects.

We create fermionic dipolar $^{23}Na^{6}Li$ molecules in their triplet ground state from an ultracold mixture of ^{23}Na and ^{6}Li. Using magnetoassociation across a narrow Feshbach resonance followed by a two-photon stimulated Raman adiabatic passage to the triplet ground state, we produce 3×10^4 ground state molecules in a spin-polarized state. We observe a lifetime of 4.6 s in an isolated molecular sample, approaching the p-wave universal rate limit. This is a 10^4- fold improvement on previous triplet lifetimes in ultracold dipolar molecules. Electron spin resonance spectroscopy of the triplet state was used to determine the hyperfine structure of this previously unobserved molecular state.



Electron spin resonance (ESR) spectroscopy. Sample spectra at B = 18.63 G showing distinct loss features for transitions in Na, NaLi, and Li. These spectra show the presence of three different species with different characteristic magnetic moments.

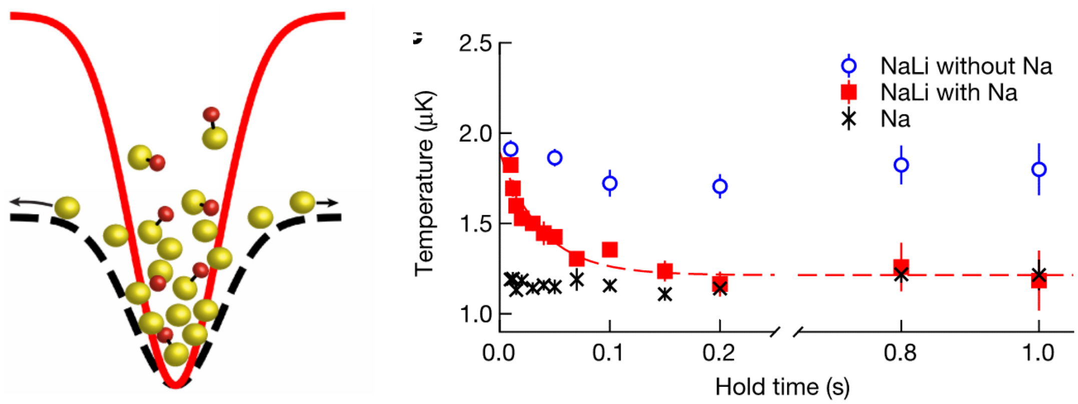
3. Collisional Cooling of Ultracold Molecules

H. Son, J.J. Park, W. Ketterle, and A.O. Jamison, *Collisional Cooling of Ultracold Molecules,* Nature **580**, 197 (2020), https://doi.org/10.1038/s41586-020-2141-z

Collisional cooling is crucial to prepare quantum degenerate gases of atoms. It has been a long-standing goal to achieve such cooling of ultracold molecules, but until now ultracold molecules could not be realized under conditions where elastic collisions were much faster than inelastic collisions. We have realized collisional cooling by sympathetically cooling rovibrational ground-state triplet NaLi molecules with Na atoms. We find a lower bound on the elastic to inelastic collision ratio greater than 50. Using two stages of evaporation, the phase-space density of the molecules is increased by a factor of 20, achieving temperatures as low as 220 nK. This is the first observation of thermalization and collisional cooling of any ultracold molecule.

NaLi is an unlikely place to find sufficiently good collisional properties since it has energetically allowed chemical reactions, even in the electronic ground state, and the triplet state we work with has an electronic excitation energy of 0.9 eV or 10, 000 K. We believe that the favorable collisional properties of the NaLi-Na mixture in fully stretched states result from strong suppression of electronic spin flips during collisions, which could otherwise lead to chemical reactions. We speculate that for NaLi, the low molecular and reduced masses play an additional major role since they result in small spin-orbit coupling and low density of states.

In the near future, an upgraded optical dipole trap should allow cooling into the quantum degenerate regime. Further, the atomic and molecular states used in this work are magnetically trappable. If molecules could be produced in a magnetic trap, sympathetic cooling with sodium should allow for the production of large samples of deeply quantum degenerate dipolar molecules. Such gases would open new areas of inquiry in quantum emulation with long-range interactions and quantum computing with ultracold molecules.

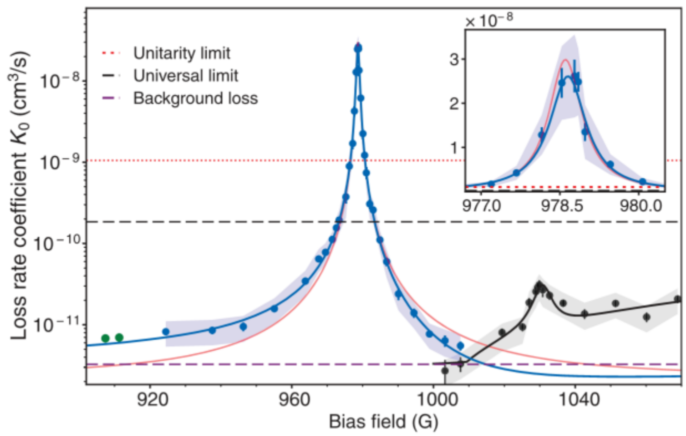


Thermalization of NaLi and Na. A quick evaporation and recompression of the trap cools the Na, but not the NaLi since the trapping potential of molecules (the red solid line) is deeper than that of atoms (the black dashed line). Subsequently, NaLi thermalizes with Na (right figure). Note that a drop in temperature does not occur in the absence of Na atoms.

4. Controlling chemistry with quantum interference: Atom-molecule Feshbach resonances

H. Son, J.J. Park, Y.-K. Lu, A.O. Jamison, T. Karman, and W. Ketterle: *Control of reactive collisions by quantum interference,* Science **375**, 1006–1010 (2022), https://doi.org/10.1126/science.abl7257

A long-standing goal in chemistry is to control reactions and collisions by external fields. We have now realized magnetic control of reactive scattering in an ultracold mixture of Na atoms and NaLi molecules via Feshbach resonances. In most molecular systems, particles form lossy collision complexes at short range with unity probability for chemical reaction or inelastic scattering leading to the so-called universal loss rate. In contrast, Na+NaLi is shown to have <4% loss probability at short range when spin polarization suppresses loss. By controlling the phase of the wavefunction via a Feshbach resonance, we modify the loss rate by more than a factor of hundred, from far below the universal limit to far above, demonstrated here for the first time. The results are explained in analogy with an optical Fabry-Perot interferometer by constructive and destructive interference of reflections at short and long range. Our work demonstrates quantum control of chemistry by magnetic fields with the full dynamic range predicted by our models. The NaLi + Na system is only the second system (besides NaK + K) for which Feshbach resonances have been observed. In ongoing work, we are characterizing such resonances over a large range of magnetic fields (1000 Gauss) and for different hyperfine states. We have found more than 30 resonances which should provide some information about collisional complexes.



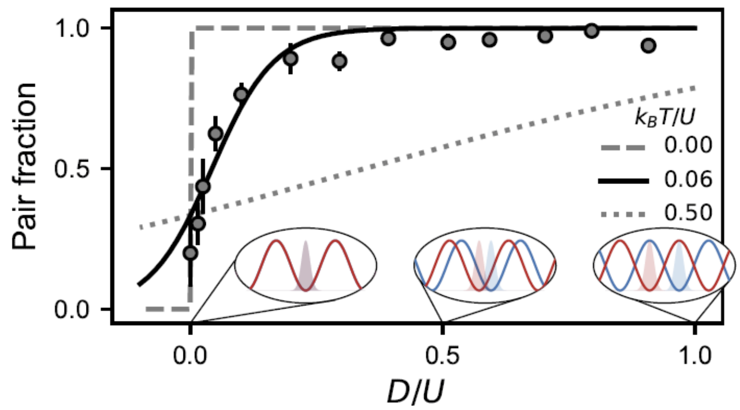
Zero-temperature loss-rate coefficients for Na + NaLi collisions. Experimental data points were corrected for nonzero momentum effects. The blue line is the best fit based on the single-channel model; the red line is a symmetric Lorentzian fit. The red dotted line is the unitarity limit in two dimensions. The black dashed line is the universal limit. The open-channel background rate (purple) is much lower than the universal limit. This allows to control collisions by multiple interference of reflections.

In our ultracold rubidium lab, we used the tunable laser for spin-dependent lattices. This allowed us to realize a new quantum state of matter, a Mott insulator of repulsively bound pairs. We had obtained preliminary results already many years earlier, but then decided to work on different projects first. The 2022 PRL is the realization of what we had proposed in the DURIP proposal.

1. A Mott insulator of repulsively bound pairs.

J. de Hond, J. Xiang, E. Cruz-Colón, W.C. Chung, W. Chen, W.C. Burton, C.J. Kennedy, and W. Ketterle: *Preparation of the spin-Mott state: a spinful Mott insulator of repulsively bound pairs.* Phys. Rev. Lett. **128**, 093401 (2022), https://doi.org/10.1103/PhysRevLett.128.093401

We observe and study a special ground state of bosons with two spin states in an optical lattice: the spin-Mott insulator, a state that consists of repulsively bound pairs which is insulating for both spin and charge transport. Two atoms in different hyperfine states are bound because their repulsive interaction is smaller than the repulsive interaction between atoms in the same hyperfine states. This is how an interaction anisotropy creates a pairing gap. Because of this gap, the spin-Mott insulator state can be prepared with low entropy and can serve as a starting point for adiabatic state preparation. We find that the stability of the spin-Mott state depends on the pairing energy, and observe two qualitatively different decay regimes, one of which exhibits protection by the gap.



Preparation of the spin-Mott state for various pairing energies D. The decrease of the pair fraction for small D is explained as the effect of finite temperature (see lines). U is the onsite repulsive energy between atoms in the same hyperfine states. The insets show the configuration of the spin-dependent lattice used for various values of D/U. Red and blue are the lattices for σ_- and σ_+ light, respectively.