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as of 08-Aug-2022

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DUNS Number: 043207562

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Final Report for Period Beginning 10-Oct-2015 and Ending 09-Mar-2019

Title: Laser Cooling and Trapping of Diatomic Molecules

Begin Performance Period: 10-Oct-2015

End Performance Period: 09-Mar-2019

Report Term: 0-Other

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

STEM Degrees: 3

STEM Participants: 7

Major Goals: The overall theme of this project was to continue developing methods for direct laser cooling and trapping of molecules. This built off our prior demonstration of the first-ever magneto-optical trap (MOT) for a molecule, using strontium monofluoride (SrF). The MOT is the workhorse technique for creating ultracold atomic gases, and one of our aims was to make the molecular MOT a similarly powerful and versatile tool. The specific major goals of this work included:

- Increase the phase-space density of SrF molecules trapped in a MOT
- Transfer SrF molecules to a conservative trap
- Study ultracold atom-molecule and/or molecule-molecule collisions using SrF
- Apply large optical forces to molecules using a novel tunable laser system

Since work began on the grant, we also opened a new line of research, with the goal to investigate the prospects for laser cooling and trapping a novel molecular species, TIF.

Accomplishments: We accomplished our first goal, by demonstrating a radio-frequency magneto-optical trap (RF-MOT) of SrF molecules with properties dramatically improved compared to prior molecular MOTs. We trapped up to $\sim 10,000$ molecules, with densities of up to $2.5 \times 10^5 / \text{cm}^3$ and temperatures as low as 250 microKelvin, with lifetimes of $\sim 1/2$ s. This corresponds to a phase-space density of nearly $10^{\{-13\}} / \text{hbar}^3$. Compared to our original molecular MOT reported by our group in 2014, this represented a 30-fold increase in the number of trapped molecules, a nearly 10,000-fold increase in phase-space density, and a 10-fold increase in trap lifetime. In an extension to this published work, we demonstrated sub-Doppler cooling of molecules from the RF MOT, reaching temperatures as low as ~ 50 microKelvin while reducing molecular density by a negligible factor. This corresponds to a ~ 25 -fold increase in phase space density. This cooling is immediately applied after molecules are released from the RF MOT, in a stage similar to the optical molasses cooling typically applied to alkali atoms from a MOT before loading into a conservative trap.

We also accomplished our second goal, by demonstrating efficient loading of molecules from the RF MOT into a conservative magnetic trap. To do this, we first applied sub-Doppler laser cooling of molecules released from the RF MOT, reaching temperatures as low as ~ 50 microKelvin while maintaining the same molecular density. We then optically pumped these molecules into a "stretched" spin state, and applied a strong magnetic field gradient to confine them, with an optimized temporal profile to guarantee trapping with minimal excess heating and, after a period of slow spatial compression, high density. To characterize this gas of trapped, ultracold molecules, we developed new techniques for diagnosing the distribution of molecular states in the magnetic trap--both the internal states, and the velocity distribution/temperature. We demonstrated loading of $\sim 4 \times 10^3$ molecules in a single

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internal state, with density $\sim 2 \times 10^5 \text{ cm}^{-3}$ and temperature $T \sim 90 \text{ } \mu\text{K}$, in the compressed magnetic trap. While this was not a sufficiently high phase space density to enable observation of ultracold molecule-molecule collisions, it should be sufficient for making initial studies of molecule-atom collisions when the trap is co-loaded with an alkali species such as Rb.

We made significant progress towards our third goal, to study ultracold molecular collisions. We constructed a 2-D MOT for creating a slow, high-flux beam of rubidium (Rb) atoms. This setup meets all mechanical constraints such that it can be attached to the main SrF experiment in the future. This should enable us to load both the RF-MOT and the magnetic trap with Rb atoms with the typical large numbers and high densities achievable with alkali atoms. This in turn should make it possible to begin studying ultracold atom-molecule collisions in the near future.

We also made significant progress towards our fourth goal, to apply large optical forces to molecules using a novel tunable laser system. The first stage of the laser system--a narrow-linewidth, long-pulse ($\sim 200 \text{ } \mu\text{s}$) laser with $\sim 2 \text{ kW}$ of instantaneous power at 532 nm --was constructed. This should be sufficient to pump a tunable laser and/or an amplifier for a cw tunable laser, as needed to implement our scheme. We also performed a numerical and theoretical study on the possibility to rapidly cool molecules (or atoms) using purely stimulated forces rather than relying on spontaneous emission. This was motivated by recent claims from the group of Hal Metcalf (Stony Brook Univ.) that cooling without spontaneous emission is possible, and indeed that it has been demonstrated using the so-called Bichromatic Force (BCF). This claim, if true, could enable spectacular advances in molecular cooling and trapping. Our work shows that these claims are not justified. Instead, we have shown that Metcalf's work is an example of a previously anticipated, but never clearly observed, mechanism for cooling. Here, the entropy associated with the velocity distribution of an ensemble of particles is traded for entropy associated with a distribution of internal states of the particles. The entropy in the internal states can then be dissipated via spontaneous emission. This mechanism is simple to quantify, and can provide cooling much more rapid than that obtained with typical laser cooling schemes (which are all based on photon scattering forces).

Finally, we worked toward a new goal, to develop methods for laser cooling and trapping of a new diatomic species, thallium monofluoride (TlF). TlF is interesting because, unlike SrF, it can be manipulated using the standard atomic method of Zeeman slowing, which should make it possible to load dramatically larger samples into a MOT. It also has closed electronic shells, and hence quite different chemical properties than SrF and all other molecular species that have been laser-cooled to date. We constructed a cryogenic molecular beam source for TlF, then used it to measure the hyperfine structure in the excited $B^3\Pi_1$ electronic state of TlF, which will be the upper state of the cycling transition to be used for laser cooling and trapping. Our analysis of the B state hyperfine structure allowed us to make quantitative predictions for limits to optical cycling on the $X^1\Sigma^+ \rightarrow B$ transition in TlF. Then, together with collaborator L. Hunter (Amherst College), we also observed optical cycling in TlF, a first for any molecule with closed electronic shells. After some optimization, we observed at least 100 photon scattering events per molecule. We developed a simple model that provides qualitative understanding of the mechanisms that limit the rate of optical cycling in species like TlF. These results constitute critical first steps on the path to laser cooling of TlF.

Training Opportunities: This grant provided support for 2 postdoctoral researchers, 5 Ph.D. students in physics, and 2 undergraduate students over its full duration. These young researchers received mentoring from the PI in all aspects of their scientific training. This sort of research exposes students to a vast array of scientific methods and concepts. They develop extensive skills and experience in areas such as numerical simulation and analysis; electronics design, construction and testing; computer-based data acquisition and analysis; laser, optical, and electro-optical operation and design; microwave and RF techniques; vacuum, gas handling and cryogenics; etc.

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Results Dissemination: In addition to the journal publications described in the Products section, our group has given many presentations about our work at conferences, workshops, and other institutions. These include the following invited talks:

- D. DeMille, AMO Seminar, University of Wisconsin, Oct. 2018
- D. DeMille, James Franck Institute Colloquium, University of Chicago, Oct. 2018
- D. DeMille, FiO/DLS "Visionary Speaker" talk, Sept. 2018
- D. DeMille, American Chemical Society Spring Meeting, March 2018
- D. DeMille, Physics Department Colloquium, UC Santa Barbara, March 2018
- D. DeMille, MIT Lincoln Laboratories Basic Science Research Seminar, Feb. 2017
- D. DeMille, Physics Department Colloquium, UCLA, Jan. 2017
- D. McCarron, American Chemical Society National Meeting, Philadelphia, Aug. 2016
- D. DeMille, International Conference on Atomic Physics, Seoul, Korea, July 2016 (Plenary)
- D. DeMille, Cold and Controlled Molecules and Ions Conference, Weizmann Institute, Israel, Mar. 2016 (Keynote)
- D. DeMille, German Physical Society (DPG) Spring meeting, Hannover, Feb. 2016 (Plenary, Keynote)
- D. DeMille, Harvard Joint Quantum Sciences Seminar, Feb. 2016
- D. DeMille, OSA Traveling Lecturer Colloquium and Seminar, Univ. of San Luis Potosi, Mexico, Nov. 2015
- D. DeMille, U.S.-Japan Seminar on Many-Body Quantum Systems, Madison, WI, Sept. 2015

as well as many contributed presentations by students and postdocs, including:

- 4 contributed presentations at the APS DAMOP 2016 meeting
- 1 contributed presentation at the APS DAMOP 2017 meeting
- 2 contributed posters at the 2017 Gordon Conference on Atomic Physics
- 1 contributed presentation and 1 contributed poster at the APS DAMOP 2018 meeting

Honors and Awards: Nothing to Report

Protocol Activity Status:

Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: PD/PI

Participant: David DeMille

Person Months Worked: 1.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Daniel McCarron

Person Months Worked: 8.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Leland Aldridge

Person Months Worked: 1.00

Project Contribution:

National Academy Member: N

Funding Support:

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Participant Type: Graduate Student (research assistant)
Participant: Matthew Steinecker
Person Months Worked: 15.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: Eric Norrgard
Person Months Worked: 9.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: Yuqi Zhu
Person Months Worked: 10.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: Eustace Edwards
Person Months Worked: 12.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
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Person Months Worked: 2.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Undergraduate Student
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Person Months Worked: 2.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Undergraduate Student
Participant: Alicia Tirone
Person Months Worked: 1.00 **Funding Support:**
Project Contribution:
National Academy Member: N

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Date Submitted: 8/31/18 12:00AM **Date Published:** 7/1/18 4:00AM
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Article Title: Magnetic Trapping of an Ultracold Gas of Polar Molecules

Authors: D.J. McCarron, M.H. Steinecker, Y. Zhu, D. DeMille

Keywords: ultracold molecules, magnetic trapping, dipolar gases

Abstract: We demonstrate the efficient transfer of molecules from a magneto-optical trap into a conservative magnetic quadrupole trap. Our scheme begins with a blue-detuned optical molasses to cool SrF molecules to $\approx 50 \mu\text{K}$. Next, we optically pump the molecules into a strongly trapped sublevel. This two-step process reliably transfers $\approx 40\%$ of the molecules initially trapped in the magneto-optical trap into a single quantum state in the magnetic trap. Once loaded, the molecule cloud is compressed by increasing the magnetic field gradient. We observe a magnetic trap lifetime of over 1 s. This opens a promising new path to study ultracold molecular collisions, and potentially to produce quantum-degenerate molecular gases via sympathetic cooling with co-trapped atoms.

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Acknowledged Federal Support: Y

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Date Received: 29-Dec-2020

Completion Date: 4/29/19 9:06PM

Title: Sub-Doppler Laser Cooling and Magnetic Trapping of SrF Molecules

Authors: Matthew Steinecker

Acknowledged Federal Support: N

Partners

I certify that the information in the report is complete and accurate:

Signature: David DeMille

Signature Date: 8/3/22 8:16PM

Nothing to report in the uploaded pdf (see accomplishments).