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14. ABSTRACT
This is the final report for our DURIP grant entitled "Apparatus for Laser Slowing and cooling of Molecules". We have successfully acquired and assembled all parts for a new cryogenic molecular beam source, and recently used the source for preliminary work on laser cooling of a new molecular species, TlF. We have also successfully acquired and assembled the parts for a custom laser system, which produces long (~200 microsecond), single-frequency pulses with energy ~1.1 Joules at 1064 nm and/or ~0.4 Joules at 532 nm. We have acquired all parts for producing tunable laser pulses based on optical parametric amplification of an external cavity diode laser with this

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Report Title

Final Report: Laser cooling and trapping of diatomic molecules

ABSTRACT

This is the final report for our DURIP grant entitled "Apparatus for Laser Slowing and cooling of Molecules". We have successfully acquired and assembled all parts for a new cryogenic molecular beam source, and recently used the source for preliminary work on laser cooling of a new molecular species, TlF. We have also successfully acquired and assembled the parts for a custom laser system, which produces long (~200 microsecond), single-frequency pulses with energy ~1.1 Joules at 1064 nm and/or ~0.4 Joules at 532 nm. We have acquired all parts for producing tunable laser pulses based on optical parametric amplification of an external cavity diode laser with this system, and assembled all but the final, yet-to-be-tested stage.

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)

Received		Paper
08/22/2013	1.00	D. DeMille, J. F. Barry, E. R. Edwards, E. B. Norrgard, M. H. Steinecker. On the transverse confinement of radiatively slowed molecular beams, Molecular Physics, (06 2013): 0. doi: 10.1080/00268976.2013.793833
08/22/2013	2.00	John F. Barry, David DeMille. Low-temperature physics: A chilling effect for molecules, Nature, (11 2012): 0. doi: 10.1038/nature11753
08/31/2014	3.00	J. F. Barry, D. J. McCarron, E. B. Norrgard, M. H. Steinecker, D. DeMille. Magneto-optical trapping of a diatomic molecule, Nature, (08 2014): 0. doi: 10.1038/nature13634
08/31/2015	5.00	D J McCarron, E B Norrgard, M H Steinecker, D DeMille. Improved magneto–optical trapping of a diatomic molecule, New Journal of Physics, (03 2015): 35014. doi: 10.1088/1367-2630/17/3/035014
TOTAL:	**4**	

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

Gordon Conference on Atomic Physics, June 2015. Poster presentation: Towards Stimulated-Force Slowing of SrF Molecules

APS DAMOP Meeting May 2016. Poster presentation, Abstract: K1.00165: Towards Stimulated-Force Slowing of SrF Molecules

Number of Presentations: 0.00

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 Book

TOTAL:

Received Book Chapter

TOTAL:

Patents Submitted

Patents Awarded

Awards

Graduate Students

NAME	PERCENT_SUPPORTED	Discipline
Eustace Edwards	0.00	
FTE Equivalent:	**0.00**	
Total Number:	**1**	

Names of Post Doctorates

NAME	PERCENT_SUPPORTED
Eric Norrgard	0.00
FTE Equivalent:	**0.00**
Total Number:	**1**

Names of Faculty Supported

NAME	PERCENT_SUPPORTED	National Academy Member
David DeMille	0.00	
FTE Equivalent:	**0.00**	
Total Number:	**1**	

Names of Under Graduate students supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

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: 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:..... 0.00

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:..... 0.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):..... 0.00

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:..... 0.00

Names of Personnel receiving masters degrees

NAME
Total Number:

Names of personnel receiving PHDs

NAME
Total Number:

Names of other research staff

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

We have successfully acquired and assembled all parts for a new cryogenic molecular beam source, and recently used the source for preliminary work on laser cooling of a new molecular species, TlF. We have also successfully acquired and assembled the parts for a custom laser system, which produces long (~200 microsecond), single-frequency pulses with energy ~1.1 Joules at 1064 nm and/or ~0.4 Joules at 532 nm. We have acquired all parts for producing tunable laser pulses based on optical parametric amplification of an external cavity diode laser with this system, and assembled all but the final, yet-to-be-tested stage.

Technology Transfer

FINAL REPORT

Year 1.

In our primary effort, we have made substantial progress towards magneto-optical trapping of SrF molecules. We have almost entirely rebuilt our previous vacuum chamber, adding several additional vacuum pumps and differential pumping regions, more optical access, and measures to reduce scattered laser light. We also set up magnetic field coils with fast current switches; several additional lasers, electro-optic and acousto-optic modulators, etc.; additional photomultipliers and a low noise CCD camera for detection; etc. We have recently used our setup to detect and optimize the flux of slow (<10 m/s) SrF molecules traversing the MOT region, as detected by the increase of their cycling fluorescence as more efficient repumping laser light is applied. We can also use the change in fluorescence due to Zeeman shifts to locate the magnetic field zero region in our CCD images. We are confident that first evidence for magneto-optic trapping of SrF will follow soon.

In the meantime, we also devised a new scheme for transversely confining molecules as they are slowed by laser light. The basic idea is to use off-resonant microwaves to apply AC Stark shifts to the molecules. By judicious choice of the microwave field profile and frequency, this should make it possible to guide slow molecules towards a trapping region with much improved efficiency (~100x or more) over our current scheme. We have constructed a prototype microwave resonant cavity for this purpose, whose properties match the calculated, expected values very well. We will install this in the existing molecular beam setup once our initial MOT studies are complete, in order to increase the number of trapped molecules.

In parallel we have begun developing a high-energy, long-pulse tunable laser system for stimulated-force slowing of molecular beams. We have obtained all of the parts for the first stage of this system (a cw-seeded Nd:YAG amplifier) and are optimizing the design to reach the specified output power from our module-based design (we are now at about 60% of spec). We have also devised a new way to use the Nd:YAG to generate tunable light, by pumping an optical parametric amplifier seeded with a tunable CW diode laser at the desired frequency. We estimate higher efficiency (and much more robust operation) than anticipated for our originally planned system (a seeded laser dye amplifier).

Year 1.

Our primary effort has been focused on the initial demonstration of a magneto-optical trap for diatomic molecules. This result, using SrF molecules, was reported in our recent paper, published in Nature. Our trap achieved the lowest temperature yet recorded for any method of directly cooling molecules. We believe this will be viewed as a major breakthrough and key enabling technology in the field of ultracold molecular physics and chemistry. Our success in creating a molecular MOT hinged on several experimental features that we have developed over the past few years. In particular, we had unusually good methods for diagnosing the velocity distribution of a slowed molecular beam, and unprecedented ability to deliver very slow molecules to a trapping region due to our development of methods for radiation-pressure slowing of the molecular beam. We also perfected methods for detecting molecules in the MOT. This included in particular the development of a technique to make extremely black, ultra-high vacuum compatible surfaces in a variety of complex shapes, as needed to suppress scattered laser light in the vicinity of the MOT. Our methods for molecular beam slowing and low-background detection are being shared with other groups in the field to speed overall progress.

Despite our success in producing the first molecular MOT, the properties of the MOT were surprising and, in some sense, a bit disappointing. We found that the trapping force was less than 1% of the usual trapping force in an atomic MOT; this in turn led to a very small number of molecules captured, at very low density, and with short lifetimes. We quickly recognized that in order to use the MOT to achieve our scientific goals (e.g. to observed novel chemical and collisional behavior of the ultracold molecules), it will be necessary to substantially improve its performance. We also developed at least a qualitative understanding of why our trapping force was so weak: given the level structure of our molecule (the same as all others now used for molecular laser cooling experiments), to first approximation calculations indicate that the force should vanish altogether. This is because, in the presence of the magnetic fields and laser polarizations needed for a MOT, the existence of dark Zeeman sublevels in the molecules should make it just as likely for them to scatter photons from the "wrong" direction (leading to an anti-

trapping force) as from the "right" direction (leading to trapping). It was, in some sense, fortuitous that there remained a tiny residual force that was nevertheless large enough to produce a detectable MOT. We have subsequently turned our attention to methods to improve the properties of our SrF MOT. We began with three plausible alternative methods, each with promise to dramatically improve our MOT trapping force. In the first, we applied chirped microwave pulses to rapidly drive population out of molecular dark states and into bright states --so that trapping forces should predominate over anti-trapping forces. Nevertheless, we saw little improvement in the MOT properties. Next, we used a non-standard configuration of optical polarizations: lin-45-lin rather than sigma+/sigma+ as is typical in a MOT. This was motivated by calculations of the optical forces on atoms with level structure similar to that of our molecules (i.e. dark Zeeman sublevels) in a 1-D geometry; for that system, the lin-45-lin configuration can give strong stimulated forces towards the MOT center while the sigma+/sigma+ configuration gives zero net restoring force. Again, nevertheless we saw little improvement from using this configuration.
Now, we have focused instead on using a time-dependent configuration of magnetic field gradients and laser polarizations. Here the B-field and polarization must be modulated synchronously, and rapidly enough (a few MHz) to non-adiabatically destabilize dark Zeeman sublevels in the molecule. Jun Ye's group at JILA has demonstrated this RF MOT principle by applying 2D magneto-optical compressive forces to the transverse motion of a molecular beam. In addition, John Doyle's group at Harvard has shown that this configuration leads to enhanced MOT formation when driving an atomic transition with dark Zeeman sublevels. The RF MOT requires substantial engineering, since the required size of RF magnetic field gradient can only be achived by dissipating 10's of watts of RF power in a resonant tank circuit inside the UHV MOT chamber. With the assistance of the Doyle and Ye groups we have completed a design of our RF MOT coils and are awaiting arrival of the parts. We are optimistic that this method will indeed yield dramatic improvement in the molecular MOT's operation.

In parallel, we have continued development of our novel, long-pulse tunable laser system with the goal of using it to apply very strong stimulated optical forces to molecules. This would circumvent the need for a strongly closed optical cycling transition in order to slow, cool, and trap molecules; this in turn would dramatically increase the range of molecular species available for these techniques. We have now succeeded in generating the 1.1J, 100 microsecond, single-frequency pulses of 1064 nm light. We have also frequency-doubled this light to produce long pulses of 532 nm single-frequency light, as desired to pump a single-frequency, cw-seeded optical parametric amplifier (OPA). At present our doubling efficiency is >30%, and we are working now to achieve the desired and anticipated 50% level. The tunable cw diode seed laser is ready to use once the 532 nm source is working at the required level; a suitable nonlinear crystal for the OPA has been identified and will be ordered once funds are available.

Year 3.
In the past year, our primary effort has been focused mainly on understanding the behavior of, and improving the performance of, our recently-demonstrated magneto-optical trap (MOT) for strontium fluoride (SrF) molecules. In the previous year, we published a paper in Nature describing first-ever molecular MOT, which at the time achieved the lowest temperature ever recorded (~2 mK) for any method of directly cooling molecules. However, despite this breakthrough, the properties of the MOT were surprising and, in some sense, a bit disappointing. We found that the trapping force was less than 1% of the usual trapping force in an atomic MOT; this in turn led to a very small number of molecules captured, at very low density, and with short lifetimes. We also developed a qualitative understanding of why our trapping force was so weak. Given a crude approximation to the level structure of our molecule (when ignoring spin substructure, a J=0 ◊ J'=1 transition, the same as all others now used for molecular laser cooling experiments), to first approximation calculations indicate that the force should vanish altogether. This is because, in the presence of the magnetic fields and laser polarizations needed for a MOT, the existence of dark Zeeman sublevels in the molecules should make it just as likely for them to scatter photons from the "wrong" direction (leading to an anti-trapping force) as from the "right" direction (leading to trapping). It hence appeared fortuitous that there remained a tiny residual force that was nevertheless large enough to produce a detectable MOT. In any case, it was clear that in order to use the MOT to achieve our long-term scientific goals (e.g. to observed novel chemical and collisional behavior of the ultracold molecules), it would be necessary to substantially improve its performance.

Just at the beginning of the current grant year, a paper was published by M. Tarbutt (Imperial College, London, UK) that provided an explanation for many of the observed properties of our molecular MOT. Tarbutt's paper explained, for the first time, the mechanisms that lead to trapping in "type-II" MOTs, where the excited state of the trapping transition has angular momentum J' = J-1 or J for a ground state with angular momentum J. Tarbutt provided both physical arguments and simple numerical calculations to justify several novel and counter-intuitive conclusions. The first of these was that a trapping force for in a standard MOT (with a DC magnetic field gradient and counterpropagating laser beams with opposite circular polarizations) arises ONLY due to the magnetic moment in the excited state; if this magnetic moment vanishes (as for the case of J'=0 mentioned above), there can be no trapping force even if the ground state has a substantial magnetic moment. The second was that the laser polarization that leads to a trapping force is NOT determined by the Zeeman shifts that bring certain sublevels into resonance; rather, the correct polarization is set by the sign of the excited-state magnetic moment, and together with the change in J-value for the transition (i.e. whether \delta J = +1, 0, or -1). Tarbutt's calculations predicted that, as observed, our original MOT should have a nonzero but very small trapping/restoring force, in fair quantitative agreement with our observed value. He also predicted that a simple change in the choice of laser polarizations used to address the various spin-rotation and hyperfine lines of our trapping transition should lead to a substantial increase in the trapping force. However, even this force is limited by the rather small value of the excited-state magnetic moment in our case (where the magnetic g-factor is g' ~ 0.1). Finally, he confirmed that—as had already been predicted in an earlier paper from the group of Jun Ye (JILA)—an even larger trapping force could be obtained by using rapid, synchronous switching of the B-field gradient and the laser polarizations. This configuration again allows a force that depends on the much larger ground state magnetic moment (where g ~ 1). However, it requires significant engineering development to apply the required large (~10 G/cm) B-field gradients switching with the required high (~few MHz) frequency. Immediately after we became aware of Tarbutt's work, we began exploring the "Improved DC" MOT using his recommended configuration of polarizations. The results of our full array of temperature, number, density, trapping force, lifetime, etc. are described in our recent paper "Improved magneto-optical trapping of a diatomic molecule", D.J. McCarron, E.B. Norrgard, M.H. Steinecker, and D. DeMille, New J. Phys. 17, 035014 (2015). Basically, we confirmed his predictions and found spring constants ~20 times larger than in our original MOT. This also—together with some technical improvements—allowed us to achieve much longer trap lifetimes (>100 ms, a 2x improvement) than originally achieved, as well as more molecules (~2x improvement) and higher density (~6x improvement). However, we also observed much higher temperatures in this configuration: ~14 mK rather than ~2 mK as in the original MOT. Hence the usual figure of merit for ultracold gases, the phase space density, was essentially unchanged. Tarbutt's model failed to predict the large difference in temperature between the two configurations. We have begun collaborating with him to try to understand and successfully model the cause; we believe it must be due to stimulated-force heating mechanisms that were not included in his model, and are likely important at the high saturation intensities where our MOTs operate.

We subsequently began to work on the rapidly-switched MOT, which we refer to as the RF (radiofrequency) MOT. This is now working routinely in our lab. As predicted, this version of the trap indeed has much improved properties, though its behavior does not seem to scale as predicted in Tarbutt's model. We are also collaborating with him to understand the behavior we see. However, a rough outline of our observations is as follows:

--it is optimal to load the trap at the highest available laser intensity, but this results in high temperatures comparable to those observed in the "Improved DC" MOT

--the trapping force in the RF MOT is remarkably insensitive to laser intensity, and can be maintained at its best value even with powers far below the nominal saturation intensity for our transition

--as the laser intensity is lowered, the temperature of molecules in the MOT drops rapidly

--by loading the MOT at high power and then ramping to lower power, it is possible to capture a large number of molecules in the MOT and then cool them to much lower temperature. Since the trapping force remains basically the same, this cooling leads to a dramatic ompression of the molecular cloud

--the behavior is insensitive to RF switching frequency over a relatively wide range (~1-2.5 MHz), but as expected reverts to DC MOT behavior at low enough frequencies (~0.1 MHz)

--because the RF magnetic field requires driving substantial current through a large inductive load (field coils), the RF circuit uses substantial voltages (up to ~4 kV). If configured incorrectly, the voltage drop

across the coils can lead to a large E-field experienced by the molecules; this in turn can lead to unwanted mixing of rotational levels via the Stark effect, and thus loss of molecules to different rotational levels during optical cycling. It is possible that, in addition to Stark mixing, Stark shifts of the molecular energy levels could be playing a role in the behavior of the RF MOT, but this is not confirmed yet.
--To date, by using the RF MOT we have improved the number (~2000, a 4-fold increase relative to the original MOT), trapping force spring constant (10-fold increase), temperature (~0.7 mK, a 3-fold improvement), and lifetime (~500 ms, a 10-fold increase). The phase space density is now ~2000 times larger than in either DC MOT. We note now that the RF MOT trap depth, temperature, and lifetime are now quite comparable to results obtained in standard atomic (type-I, DC) MOTs. Only the molecule number is significantly worse, and hence this will be an area of focus in the near future.

We are near completion of a full characterization of behavior in the RF MOT and a paper describing it. Once this is done, we will focus on improved methods for delivering more cold molecules to the MOT for loading. Once the loading is optimized, we will attempt to transfer molecules from the MOT into a conservative trap, most likely a magnetic trap to start. This would open the path to sympathetic cooling with co-trapped atoms, and in the long term possibly to creating a Bose condensate of SrF molecules.
In parallel, we have continued development of our new system where the goal is to apply very strong stimulated optical forces to molecules. This would circumvent the need for a strongly closed optical cycling transition in order to slow, cool, and trap molecules; this in turn would dramatically increase the range of molecular species available for these techniques. Here progress has been on two fronts. First, we have nearly complete construction of a new cryogenic molecular beam source to allow testing of new slowing methods and new molecular species for laser cooling. Second, we have continued our development of a novel, long-pulse tunable laser system to create the high intensity light needed for useful stimulated slowing. Starting with the 1.1J, 100 microsecond, single-frequency pulses of 1064 nm light described in last year's report, we have improved the efficiency of frequency doubling and now produce ~500 mJ of 532 nm single frequency light. This was accomplished by careful optimization of beam shape for better focusing through the nonlinear doubling crystal. This is above the target energy needed to pump a single-frequency, cw-seeded optical parametric amplifier (OPA) for the tunable light. A suitable nonlinear crystal for the OPA has been ordered, and will be tested soon. We have already made preliminary tests of damage threshold for the relevant crystal (MgO:PPSLT) when subjected to our unusual long and high-energy pulses.

Final.
We have successfully acquired and assembled all parts for a new cryogenic molecular beam source, and recently used the source for preliminary work on laser cooling of a new molecular species, TlF. We have also successfully acquired and assembled the parts for a custom laser system, which produces long (~200 microsecond), single-frequency pulses with energy ~1.1 Joules at 1064 nm and/or ~0.4 Joules at 532 nm. We have acquired all parts for producing tunable laser pulses based on optical parametric amplification of an external cavity diode laser with this system, and assembled all but the final, yet-to-be tested stage.