Center for Astrophysics| Harvard & Smithsonian
ITAMP
60 Garden Street
Cambridge, MA 02138 USA

Laser Cooling of Molecules

October 27 - 29, 2022

Organizers

Lawrence Cheuk *(Princeton)*, **David DeMille** *(Chicago)*
John Doyle *(Harvard)*, **Tanya Zelevinsky** *(Columbia)*

Phillips Auditorium
Center for Astrophysics|Harvard & Smithsonian
60 Garden St. Cambridge, MA 02138

Instructions for Reimbursement Forms

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Please be patient as the reimbursement setup and payment process may take some time. Thank you

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Bus to Harvard Square to MIT is Bus #1
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Harvard Square to Airport: Red Line Train to South Station. Take the Silver line to Logan Airport. Silver line stops at all terminals. To see the schedule of buses visit http://www.mbta.com/schedules_and_maps/bus/

Taxicabs
Ambassador: 617 492-1100 Yellow: 617 547-3000

Dining around ITAMP

Near the Observatory	Sarah's Market, 200 Concord Ave.
	The Village Kitchen, 359 Huron Ave.
	House of Chang, 282 Concord Ave.
	Trattoria Pulcinella, 147 Huron Ave.
	Armanado's Pizza, 163 Huron Ave.
	Hi-Rise Bread CO, 208 Concord Ave.
	Formaggio Kitchen, 244 Huron Ave
Massachusetts Ave.	Chang-Sho, 1712 Mass. Ave.
	Simons Coffee House, 1736 Mass. Ave.
	Nirvana The Taste of India 1680 Mass Ave
	Stone Hearth Pizza Co., 1782 Mass.Ave.
	Super Fusion, 1759 Mass. Ave.
	Cambridge Common, 1667 Mass Ave.
	Lizard Lounge, 1667 Mass. Ave.

ITAMP began life in 1989 at the Harvard-Smithsonian Center for Astrophysics. It is the only theoretical AMO "user facility" in the United States. It hosts workshops (3-days), and visiting fellows (short- and long-term), sponsors a flagship speaker series, the Joint Quantum Science seminars, with HQI in the Physics Department, and a rigorous postdoctoral program. ITAMP workshops are recorded and avaliable on the ITAMP YouTube channel. There are on average 4-5 workshops each year. A Call for Proposal to organize workshops and a list of workshops are available at http://itamp.harvard.edu. The postdoctoral program has been a recognized success, placing energetic fellows into junior positions at universities and national labs.

ITAMP thrives in the larger Cambridge-area AMO physics ecosystem, drawing upon the considerable depth and breadth of experimental expertise. The mission of ITAMP continues to be in furthering the cause of theoretical AMO physics by providing resources, centrality of location, and scientific and administrative expertise, to enhance collaborative efforts between theory and experiment, and to be broad in advocating for theoretical AMO.

H. Sadeghpour

Laser cooling is a key technique for quantum control, since it allows trapping samples of particles at microkelvin temperatures. Hence, direct laser cooling of molecules has become a general and powerful technique. It is poised to enable production of quantum-degenerate molecular gases and large arrays of single molecules for applications in ultracold chemistry, high-precision measurement and sensing, and quantum simulation & computation. Further advances will require continued development of new experimental techniques as well as support from theorists to understand and control relevant molecular structure and collisions.

Thus, an important goal of the proposed workshop is to create a sense of community and collaboration for the increasing number of molecular laser cooling groups, and to provide a venue for dynamic discussions.

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Welcome to ITAMP

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60 Garden St.
Cambridge, MA 02138

Who to Contact:

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Internet Access
Most wireless devices will connect to the “Harvard Guest” account. This works fine and there is no paperwork involved in getting internet access. Other connections such as edurom are available.

Copiers/Faxing and Phone Access
Copy Machines are located throughout the building. Should you need access to a copier please see someone at ITAMP.

Telephone System
To call outside the University you must dial 9 before the number. The University prefix three digits are 495 and 496. To dial on campus, you simply need to dial the “5” or “6” and the last four digits. For example, ITAMP Admin office’s number is 617-495-9524. You can dial 5-9524 to call internally. The Institute is not permitted to pay for long distance calls.

Pawel Wojcik

University of Southern California

““Quantum chemistry for quantum information science”

During the last decade the complexity of laser cooled molecules has rapidly increased. Description of polyatomic species requires use of quantum chemistry programs. Wide access to- and awareness of methods offered by programs like Q-Chem promises progress in the field. I will present tools of computational chemistry used in our group with a focus on applications to laser cooling of molecules. The family of equation-of-motion coupled-cluster (EOM-CC) methods is our main device used to describe the electronic structure and related properties of the systems that we considered for optical cycling. Additionally, I will present ways of visually analyzing electronic wavefunctions with natural transition orbitals (NTOs) and Dyson orbitals which are helpful in interpretation and design of desired molecular properties. I will cover tools that our group uses to calculate Franck-Condon factors (FCFs) in a process of resolving the vibronic structure. I will conclude by presenting recent results from our group that focused on laser cooling of molecules. https://iopenshell.usc.edu/research/qis/

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Durham

"An update on ultracold molecules theory and experiment at Durham University"

I will discuss a python package I have been working on to calculate the quantized rotational and hyperfine structure of 2-Sigma molecules in the presence of external fields. This work is based on the diatomic-py module recently published for 1-Sigma molecules (arXiv:2205.05686v1). I will also give an update on the status of the RbCs tweezer experiment.

Timur Tscherbul

UN Reno

"Magnetic tuning of electric dipolar interactions in ultracold gases of open-shell molecules: Feshbach resonances and avoided crossings"

Recently, a single, pronounced magnetic Feshbach resonance has been observed in ultracold collisions of triplet ground-state NaLi molecules at a magnetic field, where two open hyperfine-Zeeman states of NaLi become nearly degenerate [1]. I will discuss a theoretical model of this resonance, which postulates a p-wave bound state coupled to two open channels. The model predicts a remarkable increase of the inelastic loss rate with narrowing the energy gap between the open channels. This unexpected new type of degeneracy-induced Feshbach could be ubiquitous in ultracold molecular physics, offering a powerful new mechanism for tuning intermolecular interactions with external electromagnetic fields.

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Stefan Truppe

Imperial/FHI

"Laser Cooling in the Deep Ultraviolet - Challenges and Opportunities"

We present our recent progress on laser cooling AlF molecules using deep UV lasers. AlF has very diagonal Franck-Condon factors and its electronic structure resembles that of alkaline earth metal atoms and Zn and Cd. AlF has a strong transition near 227.5 nm for rapid slowing into a magneto-optical trap with a large capture velocity. It has also a narrow, spin-forbidden transition near 367 nm for high-resolution spectroscopy and narrow-line cooling. The large transition-linewidth of 84 MHz, the small ground-state g-factors and the inherent stability of the deeply bound AlF molecule allow loading a MOT from a continuously operating thermochemical source by using a simple, permanent-magnet Zeeman slower. Narrow-line or sub-Doppler cooling to uK temperatures can then be applied to increase the phase-space density. At high densities, chemical reactions of the molecules are suppressed because the bimolecular reaction channel is strongly endothermic.
We show that rapid optical cycling on any Q line of the transition is indeed possible, despite the unresolved hyperfine structure in the ground-state. Record-high optical scattering rates are achieved without the need for rf-sidebands or polarisation modulation. To assess if sub-Doppler cooling can be implemented, we reduce the transverse velocity distribution of a cryogenic molecular beam by using a blue-detuned molasses. To study the feasibility for narrow-line cooling, we measure the lifetimes of the levels. This allows us to present a scheme to tune the effective linewidth of the narrow-line by simultaneously driving the two spin-forbidden and transitions. High-power deep UV lasers are required to saturate the strong laser cooling transition. To address this challenge, we develop stable laser systems with a small footprint and explore new geometries for doubling-
cavities to reduce UV-induced damage in the non-linear crystal. To test these lasers and the AlF MOT apparatus, we load a MOT of Cd atoms from a cryogenic buffer gas beam and reach densities close to 10^{12} cm^{-3} .Cd is not only an excellent dummy atom to test our AlF apparatus but is an excellent candidate to search for new physics beyond the Standard Model by using high-precision isotope shift spectroscopy. We present first experimental results to benchmark new atomic structure calculations and show that kHz-level narrow-line spectroscopy in Cd can be a very sensitive probe for new physics. High-power deep UV lasers are required to saturate the strong laser cooling transition. To address this challenge, we develop stable laser systems with a small footprint and explore new geometries for doubling cavities to reduce UV-induced damage in the non-linear crystal. To test these lasers and the AlF MOT apparatus, we load a MOT of Cd atoms from a cryogenic buffer gas beam and reach densities close to 10^{12} cm^{-3} . Cd is not only an excellent dummy atom to test our AlF apparatus but is an excellent candidate to search for new physics beyond the Standard Model by using high-precision isotope shift spectroscopy. We present first experimental results to benchmark new atomic structure calculations and show that kHz-level narrow-line spectroscopy in Cd can be a very sensitive probe for new physics.

Mike Tarbutt

Imperial

"Collisions between laser-cooled molecules and atoms"

The study of collisions between ultracold atoms and molecules is important for advancing our understanding of these fundamental processes. Such collisions will be useful for sympathetic cooling of molecules, formation of ultracold polyatomic molecules, and the control of interactions through Feshbach resonances. We make mixtures of ultracold CaF and Rb and have studied their collisions in magneto-optical traps, magnetic traps, and optical traps. I will discuss our progress so far and future opportunities.

Laser Cooling of Molecules

October 27 - October 29, 2022
Phillips Auditorium

Thursday, October 27, 2022

Chair: Tanya Zelevinsky

8:00 - 8:15 am	Registration - Coffee & Pastries
8:15 - 8:30 am	Welcome and Introduction
8:30 - 9:00 am	Thomas Langin
9:00 - 9:30 am	Justin Burau
9:30 - 10:00 am	Stefan Truppe
10:00 - 10:30 am	Break
10:30 - 10:50 am	Boerge Hemmerling
10:50 - 11:20 am	Daniel McCarron
11:20 - 11:40 am	Micro Siercke
12:00 - 2:00 pm	Lunch

Chair: John Doyle

2:00 - 2:20 pm	Eunmi Chae
2:20 - 2:50 pm	Mike Tarbutt
2:50 - 3:20 pm	Matthew Frye
3:20 - 3:40 pm	Tijs Karman
3:40 - 4:00 pm	John Bohn
4:00 - 4:30 pm	Break
4:30 - 5:00 pm	Lawrence Cheuk
5:00 - 5:30 pm	Timur Tscherbul
5:30 - 6:00 pm	Tim Langen

7:00 pm Dinner Physics Library, 4th floor of Jefferson Building

Friday, October 28, 2022

Chair: David DeMille

8:00 - 8:30 am	Refreshments
8:30 - 9:00 am	Loic Anderegg
9:00 - 9:30 am	Ian Lane
9:30 - 10:00 am	Debayan Mitra
10:00 - 10:30 am	Break
10:30 - 10:50 am	Hannah Williams
10:50 - 11:20 am	Svetlana Kotochigova
11:20 - 11:40 am	Pawel Wojcik
12:00 - 1:30 am	Lunch

Chair: Lawrence Cheuk

1:30 - 2:00 pm	Gerhard Rempe
2:00 - 2:30 pm	Lan Cheng
2:30 - 3:00 pm	Jinjun Liu
3:00 - 3:30 pm	Ben Augenbraun
3:30 - 4:00 pm	Break
4:00 - 4:20 pm	Claire Dickerson
4:20 - 4:40 pm	Wes Campbell
4:40 - 5:00 pm	Eric Hudson
5:00 - 5:20 pm	Yuki Miyamoto

Saturday, October 29, 2022

Chair: Hossein Sadeghpour

8:00 - 8:30 am	Refreshments
8:30 - 9:00 am	Nick Hutzler
9:00 - 9:20 am	Zack Lasner
9:20 - 9:50 am	Nathan Clayburn
9:50 - 10:20 am	Break
10:20 - 10:50 am	Jongseok Lim
10:50 - 11:20 am	Eric Norrgard
11:20 - 11:50 am	Robert Berger
12:00 pm	Adjourn

Mirco Siercke

Hannover

"Progress on Zeeman slowing of CaF"

Techniques to cool and manipulate molecules are making incredibly rapid progress. Measurements of fundamental physics, ultracold chemistry and quantum computation have all been demonstrated in just the last few years. As these experiments become more and more complex, the system size also becomes more important, and many experiments stand to gain from an increase in the initial number of particles. To accomplish such an increase, we have set out to realize Zeeman slowing of CaF molecules, a technique that should be able to both compress the velocity distribution of a molecular beam and slow it down continuously [1][2]. In this talk, I will show our data measuring the Zeeman force directly in a proof-of-principle experiment [3], as well as our most recent slowing results. I will also talk about our ideas for improving current molecular MOTs by engineering the sub-Doppler forces inherently present in these systems [4].

Gerhard Rempe

MPQ

"Cold collision experiments with trapped polyatomic molecules"

Understanding the world around us requires exploring molecules and their interaction with other molecules. Towards this goal, we have developed several new cooling and trapping techniques, all utilizing the permanent electric dipole moment of polyatomic molecules. These techniques include electrostatic skimming, guiding and trapping, centrifuge deceleration as well as cryogenic buffer-gas and Sisyphus cooling. Combined in one setting, we use them to prepare samples of simultaneously cold, dense, and slow H_2CO and CH_3F molecules for high-resolution spectroscopy and controlled-collision studies, respectively. Our methods open up new possibilities for a plethora of experiments in various research fields ranging from cold chemistry to quantum information.

Loic Anderegg

Harvard University

"Laser Cooling of CaF and CaOH"

Ultracold molecules have a wide range of potential applications spanning from fundamental physics to quantum simulation and computation. Motivated by potential discoveries in these areas, significant advances in controlling molecules at the single-quantum-state level have occurred over the past decade. Progress in direct laser-cooling of molecules has led to the first molecular magneto-optical traps, which have allowed for optical trapping of ultracold molecules. Optical tweezer arrays have permitted both high-fidelity readout as well as quantum control of individual molecules. In this talk, we will discuss using optical tweezer arrays of CaF molecules to study ultracold collisions and quantum bits based on rotational states. Collisions are studied through trap loss and controlled by applying microwave radiation to directly engineer and tune the interaction potentials between molecules. With specific microwave detuning, a repulsive shield is created that suppresses inelastic loss, a generalizable approach to applying evaporative cooling to molecules. We also demonstrate greatly improved rotational coherence times for molecular qubits in optical tweezer traps, which parametrizes the potential performance of polar-molecule-based quantum simulators or computers. We will show progress we are making towards realizing the goal of high-fidelity molecular qubits with new, larger CaF arrays, and demonstrate dipolar interactions between CaF molecules. Extending laser cooling beyond the rich structure of diatomic molecules to polyatomic molecules leads to powerful new scientific avenues. In particular, the presence of closely spaced opposite parity levels offers a new frontier in quantum science. We will present results on the laser-cooling and optical trapping of polyatomic CaOH molecules. Recent results on manipulation of parity doublet states will also be presented. Finally, we will present electron spin precession using optically trapped CaOH molecules, a roadmap to future electron electric dipole moment searches.

Ben Augenbraun

Harvard University

"Cooling Asymmetric Tops and Large Molecules"

The diversity of electronic structures, geometries, and atomic constituents present in polyatomic molecules potentially makes them powerful building blocks for next-generation experiments in quantum science and precision measurement. Using complex molecules for these applications requires us to confront one of their most defining characteristics: molecular asymmetry. Until recently, the lack of strict selection rules was thought to make infeasible the laser-based control of asymmetric top molecules (ATMs; molecules with three distinct moments of inertia). We have shown that this is likely not the case and have put forward an experimental toolbox for optical cycling and laser cooling of ATMs. In addition, we have proposed molecular design principles that allow pushing beyond the M-O-R motif that is characteristic of the polyatomic molecules that have so far been laser cooled—opening up a wider world of unusual geometries and ro-vibrational modes for scientific use. Experimental studies of the molecules CaSH, $CaNH_2$, and $CaOC_6H_5$ validate key findings of our theoretical results and demonstrate initial progress toward optical cycling and laser cooling of ATMs.

Eric Norrgard

NIST

"Blackbody Radiation Metrology with Laser Cooled Molecules"

Polar molecules are intrinsically sensitive sensors to electric fields, including blackbody radiation (BBR). In fact, BBR has been determined to limit the trap lifetime of laser cooled polar molecules in some recent experiments. The National Institute of Standards and Technology (NIST) uses well characterized blackbody cavities as reference standards for radiometric calibrations of devices ranging from photodiodes to bolometers to cameras. I will describe NIST's recent efforts to use sensitive, identical quantum systems (such as polar molecules), to directly measure the blackbody electric field of reference blackbody cavities in order to better characterize these calibration standards. I will also present recent measurements of the radiative lifetime and Franck-Condon factors for the A state of the MgF molecule, which we have chosen as the laser cooled molecule for our BBR sensor.

Yuki Miyamoto

Okayama

"High resolution spectroscopy of buffer gas-cooled phthalocyanine"

We have been studying high-resolution electronic spectroscopy of buffer gas-cooled molecules. Recently, we measured spectra of a relatively large molecule, phthalocyanine. The spectra show an oscillation-like pattern that is attributed to the rotational structure. Analysis of this spectrum revealed that both the translational and rotational temperatures are below 10 K. The fact that the spectrum of phthalocyanine cooled by neon buffer gas is similar to that cooled by helium suggests that the spectrum we measured is not due to clusters with buffer gas atoms but to isolated molecules. In my talk I would like to present our recent results.

Robert Berger

Phillips Uni. Marburg

"Opportunities through highly-charged molecules"

In this presentation, I will discuss the opportunities that cold molecular highly-charged ions containing actinides can provide in the search for violations of fundamental symmetries. As one of the specific examples, I will highlight the case of PaF^{3+}, which we predicted in Ref. [1] to show high sensitivity for a pronounced Schiff moment of the ^{229}Pa nucleus, and will address prospects to obtain laser-cooled molecular highly-charged ions.

John Bohn

JILA

"Slow as Molasses: Viscosity in an Ultracold Dipolar Gas"

The physics of quantum degenerate dipolar gases, for example supersolid BECs, is rich and has been widely studied. By contrast, ultracold thermal gases just outside of quantum degeneracy remain a wide-open area of investigation. To kick off the discussion of this topic, we have developed the fluid mechanics equations for such gases in the hydrodynamic regime, including heat conduction and viscosity tensors, both of which display anisotropy inherited from collisions of the polarized constituents. These equations are meant to describe phenomena in ultracold gases composed of dipolar molecules, produced by laser cooling or other means.

Debayan Mitra

Columbia

"Laser cooling of CaH - mitigation and control of predissociation"

Recent progress in the laser cooling of diatomic, triatomic and symmetric top molecules have laid out favorable properties that make some molecules amenable to repeated photon scattering. However, sometimes a molecule is desired for certain properties that inhibit or impede this optical cycling. One such molecule is calcium hydride. This molecule is attractive as a pathway to obtain ultracold and trapped samples of atomic hydrogen. However it suffers from the problem of predissociation. In the first part of my talk, I will describe how we can identify useful transitions and scatter enough photons to demonstrate laser cooling. In the second part, I will discuss how we experimentally probe the predissociation probability of the B state of this molecule. I will also share theoretical insights on predissociation and show that its effect can be mitigated. I will conclude by showing how we could harness this natural ability of the molecule to break apart to perform a controlled dissociation leading to cold atomic hydrogen.

Daniel McCarron

UConn

"Laser-cooled molecules for quantum science and controlled organic chemistry"

Laser-cooling and trapping techniques for molecules promise access to a diverse range of ultracold species for applications such as quantum simulation and improved precision measurements. In this talk, I will present recent progress from two experiments within my group to laser-cool and trap two different species for complementary studies of molecule-molecule interactions. The first, AlCl, has an electronic structure analogous to that of alkaline earth atoms and presents a number of advantages for realizing large, dense samples of laser-cooled molecules. The second, CH, has traditionally been a challenging molecule to produce at high density but offers access to studies of simple controlled organic chemistry that may be compared to calculations by quantum chemists.

Justin Burau

JILA

"Reaching a new cooling limit with YO molecules"

Ultracold molecules hold promise for many important applications, ranging from quantum simulations and quantum information processing to precision tests of fundamental physics. Over the past decade, direct laser cooling of molecules has successfully achieved magneto-optical trapping, cooling to sub-Doppler temperatures, and loading into conservative traps. Achieving quantum degeneracy of directly laser cooled molecules remains an important goal for opening many applications for fundamental studies. Our group has recently demonstrated the highest phase-space density [3x10-6] for a bulk gas of dipolar molecules in a lattice trap [1]. However, the low transfer efficiency into the conservative traps prevents serious attempts towards quantum degeneracy, owing to the small number of molecules and large cloud sizes.

In this talk, we present our recent progress in this frontier with YO molecules. We have demonstrated the first molecule-based, Blue-detuned Magneto-Optical Trap [2]. We achieve sub-doppler temperatures and small cloud sizes, that cumulatively increase the number density of YO by about an order of magnitude, compared to our previous best in free space [3]. Molecules released from the Blue-detuned MOT are rapidly cooled further down to a few microkelvins. This step is crucial for our next goal of achieving near unity transfer efficiency into a conservative optical trap. I will also briefly describe our progress in building a narrowline MOT, akin to those for alkaline earth atoms, with the same goal towards achieving quantum degeneracy for YO.

Wes Campbell

UCLA

“Laser cooling mechanism and generalization to molecular rotation”

The energy-selective step in Doppler cooling comes from the Doppler effect. Following a derivation by Fermi of the translational Doppler effect, I will present the rotational Doppler effect and describe how this may allow laser cooling of molecular rotation even under conditions of poor band resolution. Since the energy-selective step in Doppler cooling can be reversible, I will also discuss some counter-intuitive conclusions about the light driving the actual cooling.

Jinjun Liu

University of Louisville

“Spin-vibronic interactions in asymmetric-top open-shell molecules and their effects on vibrational and rotational branching ratios”

Vibrational branching ratios (VBRs, closely related to Franck-Condon factors) and rotational branching ratios (RBRs, closely related to Hönl–London factors) are crucial to direct laser cooling of molecules. For asymmetric-top open-shell molecules, spin-vibronic interactions, e.g., the spin-orbit (SO) interaction, the Jahn-Teller (JT) interaction, and the pseudo-Jahn-Teller (pJT) interaction, and related effects can all alter the vibrational transition intensities and, hence, the VBRs. For quasi-degenerate electronic states, the RBRs are also affected due to the mixing of electronic states by SO and Coriolis interactions. I will present results from recent work [1-4] on spin-vibronic interactions in asymmetric-top molecules by our group and collaborators. The
ongoing effort in our lab toward accurate measurement and prediction of VBRs and RBRs will be discussed briefly.

Jongseok Lim

Imperial

"Laser-cooled molecules for tests of fundamental physics"

Despite its many successes, the Standard Model of particle physics is thought to be incomplete, because it leaves unanswered several major questions. One of these is the origin of the observed asymmetry between the amount of matter and antimatter in the visible universe. While we cannot currently explain what caused the asymmetry, we know that it requires the presence of new interactions violating several fundamental symmetries. Electric dipole moments of elementary particles are one direct signature of such symmetry violation, which can be tested via precision measurements using heavy polar molecules. In this talk, I will discuss how we plan to make extremely precise measurements of the electron electric dipole moment using molecules cooled to microkelvin temperatures. With very careful measurements, such table-top experiments will enable us to explore new physics up to PeV energy scale.

Eunmi Chae

Korea University

"Towards laser cooling of MgF"

Due to their complex internal structures and strong long-range interactions, diatomic molecules are expected to be a promising platform for quantum simulations/computations. An ideal quantum platform should be able 1. to manipulate every qubit with perfect freedom and 2. to entangle any sets of qubits in the system. The rotational states of the molecules trapped in an optical tweezer array form qubit states with long coherence time. And both global and local addressing of the qubits can be easily implemented by adjusting the trap depth or polarization of the individual tweezers.

Here, we introduce a way to generate entanglement between molecules regardless of their positions by coupling them through a microwave cavity. As optical cavities, a microwave cavity can entangle molecules in its mode volume when its resonant frequency equals to the rotational transition frequency of the molecules. One can select which molecules to couple by adjusting the transition frequency of each molecule using the tweezers. More detailed scheme of the entanglement and its feasibilities will be discussed in the presentation. At the last part f the talk, I will also briefly share the progress of a new MgF experiment at Korea University.

Lan Cheng

Princeton

“Vibronic-structure calculations for laser-coolable polyatomic molecules”

The presentation will be focused on the development and application of relativistic quantum-mechanical methods for accurate electronic- and vibrational-structure calculations for laser-coolable molecules. Intensity borrowing mechanisms for nominally symmetry-forbidden transitions in linear triatomic molecules via spin- vibronic coupling perturbations are discussed with example calculations for CaOH and a prediction for RaOH. Calculations of lifetimes for vibrational ground and excited states are also discussed using ThF^+ and CaOH as examples.

Zack Lasner

Harvard University

“SrOH”

SrOH was the first molecule to be laser-cooled, using only one repumping laser. Recent high-resolution vibrational branching ratio measurements show that its optical cycle could be further closed to make magneto-optical trapping feasible with 8—10 lasers, fewer than currently used to trap CaOH. This will enable measurements with large numbers of SrOH molecules in long-lived states and a highly controlled environment for precision measurements. I will discuss two immediate applications of laser-cooled SrOH molecules. First, the rich structure of polyatomic molecules promises robustness against systematic errors in a search for the electron electric dipole moment, while the relatively heavy Sr nucleus provides sensitivity to CP-violating interactions. Second, the multiplicity of distinct vibrational modes enables precision rovibrational microwave spectroscopy to probe certain dark matter models via measurements of the time-dependence of the proton-to-electron mass ratio. Experimental progress toward trapping SrOH is discussed.

Thomas Langin

University of Chicago

"Polarization enhanced cooling of SrF Molecules in an Optical Dipole Trap & New Techniques for Improved Molecular Magneto-Optical Trapping"

Reaching quantum degeneracy in 'directly' cooled and trapped molecular systems seems increasingly within reach. One remaining roadblock on this path are the low molecular densities and phase-space densities achieved in bulk trapping of molecules in optical dipole traps [1, 2, 3, 4, 5]. Higher density will allow for evaporative cooling to occur on a timescale fast enough to enable evaporation in the presence of various loss mechanisms. We have demonstrated that, by choosing optimal polarizations of the ODT laser and of intensity imbalanced counter-propagating Λ-enhanced gray molasses lasers, SrF molecules loaded into an ODT from a magneto-optical trap (MOT) can be cooled to temperatures as low as 14μK in an ODT with trap depth 570μK. Tuning the Λ light intensity balance yields a 10x (100x) increase in trap density (phase space density), for the ideal polarization choice. We also report on simulations of new techniques to improve the MOT spatial and phase-space density. These include simulations of methods to increase the flux of slow molecules delivered to a MOT region; two-color red-detuned MOTs with improved capture velocity; and two-color blue-detuned MOTs to very effectively cool and compress already captured molecules. Altogether, these methods hold the promise to improve the density prior to ODT loading by a factor of 103 or more.

Lawrence Cheuk

Princeton

"On-Demand Entanglement of Laser-cooled CaF Molecules in a Reconfigurable Optical Tweezer Array"

Entanglement is crucial to many quantum applications including quantum information processing, simulation of quantum many-body systems, and quantum-enhanced sensing. Although molecules have long been proposed as a promising platform for quantum science, deterministic entanglement of individually controlled molecules has been a long-standing experimental challenge.

In this talk, I will present our recent work on entangling laser-cooled CaF molecules on-demand. Using the electric dipolar interaction between pairs of individual molecules prepared using a reconfigurable optical tweezer array, we realize an entangling two-qubit gate that is sufficient for universal quantum computing and use it to deterministically create Bell pairs. Our results demonstrate the key building blocks needed for quantum information processing, simulation of quantum spin models, and quantum-enhanced sensing. They also open up new possibilities such as using trapped molecules for quantum-enhanced fundamental physics tests and exploring collisions and chemical reactions with entangled matter.

Nathan Clayburn

Amherst

"Optical Cycling of Thallium Fluoride"

Thallium fluoride molecules are a promising candidate for measurements of the thallium nuclear Schiff moment. A non-zero Schiff moment would imply a violation of time-reversal symmetry and could improve our understanding of the cosmological matter-antimatter asymmetry. To obtain the desired experimental sensitivity, sufficient optical cycling needs to be achieved so that state detection can be accomplished with near unit-efficiency. We performed detailed optical cycling studies by laser exciting a cryogenic molecular beam and recording the resulting UV fluorescence. Simple theoretical models fail to describe these experiments as hyperfine dark states of the ground state are found to dramatically reduce photon cycling rates. More complete simulations which solve the optical Bloch equations and track trajectories are in qualitative agreement with our experimental measurements. With the aid of these simulations, we have achieved sufficient optical cycling to allow near unit-efficiency detection. Further improvement in the optical cycling remains under investigation with the hope of achieving transverse cooling of the molecular beam.

Tim Langen

Struttgart

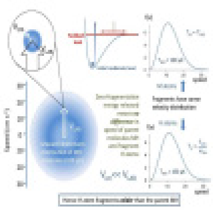
"Laser cooling of BaF molecules, and why you should care about molecular Bose-Einstein condensates"

In the first part of my talk I will report on our progress towards laser cooling of BaF molecules. Due to its high mass, resolved hyperfine structure in the excited state and branching losses through intermediate states, this molecular species is notoriously difficult to cool, but it shows high promise for various types of precision measurement applications. In the second part, I will discuss the new possibilities that bulk molecular Bose-Einstein condensates may open up for dipolar many-body physics in the near future. Building on our work on dipolar droplets and supersolids that form from weakly dipolar atoms, I will show how ultracold molecules and microwave shielding can provide fundamentally new insights into these exotic states of matter.

Ian Lane

Belfast

"Breaking BaD"



Laser cooling can produce gaseous environments unlike any other technology, reaching temperatures beyond conventional cryogenics where wave-like behaviour is dominant. Recently, the method has been applied to molecules as well as atoms but the increased complexity is significant. This talk will discuss the application of laser-cooled molecules as a source of ultracold fragments, such as carbon atoms and the halogens like F, that are challenging to directly cool. We will follow the evolution of the method to one aiming to produce extremely low temperatures (see scheme above) in atoms of low mass, particularly for the case of hydrogen. The challenges of cooling BaH and BaD and how to accurately model the laser cooling process are presented, such as how changes in vibrational spacing and rotational constants subtly effect the cooling dynamics and ultimately the final temperatures. The potential applications for ultracold hydrogen and deuterium are briefly touched upon.

Claire Dickerson

UCLA

"Molecular design for laser coolable molecules"

Electronic excitations, induced by lasers, that decay directly to the initial electronic and vibrational state are called optical cycling transitions. These transitions are used in quantum information and precision measurement for state initialization and readout. We will discuss various design rules from chemical principles for ultranarrow molecular electronic transitions of increasingly large molecules. Additionally, we will touch on the theoretical techniques to quantify good optical cycling center candidates and match our results to experiment.

Matthew Frye

Warsaw/Durham

"Modelling collisional loss of Ultracold Molecules"

Collisions are of crucial importance in understanding and controlling ultracold molecular gasses. However, they come with the potential for loss of molecules - whether by inelastic, reactive, or other loss pathways - which has been both a key challenge to the progress of many ultracold molecules experiments, and a probe into the physics of atom+molecule and molecule+molecule interactions. However, full theoretical description of such collisions is prohibitively complicated and computationally expensive for most systems of interest, which has driven work on approximate methods to capture the important physics.

In this talk I will present the methods we have used to study collisional loss in ultracold collisions, which are based on quantum defect theory using simplified long-range interactions and use partially absorbing (or capture) boundary conditions. I will discuss the recent experiments they have been applied to, covering over 3 orders of magnitude in temperature, and what can be learned from them. Finally, I will talk about our work to extend this method to arbitrarily complicated interactions and boundary conditions, with a focus on modelling collisional shielding with static electric fields.

Abstracts

Svetlana Kotochigova

Temple

"Laser Coolable Magnetic Hybrids"

The interactions of alkaline-earth atoms with inorganic molecules is a fascinating subject with a variety of applications. Of particular interest in the context of laser cooling are alkaline-earth monoxide molecules with their chemical bond that is dominated by a single-valence electron. These molecules are both polar and paramagnetic and thus can be manipulated by electric and magnetic fields as well as laser cooled. An optically-trapped array of such cold molecules is a candidate for error-free quantum computation.

Here, we investigate the likelihood of a) direct laser cooling of the highly magnetic rare-earth (RE) monoxide molecules and b) attachment of an optical cycling center (OCC) to functionalized fullerenes, i.e C60 and C80, with imbedded rare-earth atoms as a platforms to form a unique quantum computer architecture. For both systems, complete and accurate information on the potential energy surfaces and magnetic properties was previously unavailable. The magnetic RE atoms have inner shells of electrons that remain unfilled while electrons occupying outer shells participate in chemical bonding. In fact, the electrons of the partially occupied inner shells with a number of their spins aligned are responsible for the magnetic and anisotropic properties of RE molecules. The detailed study of these complex systems will allow us to theoretically identify the key properties for their direct laser cooling.

Tijs Karman

Radboud University

"Symmetry breaking in sticky collisions"

Ultracold molecules undergo "sticky collisions" that result in loss even for chemically nonreactive molecules. Our current best theory of this process is statistical RRKM theory, which shows that the sticking time is proportional to the density of states of the collision complex. Hence, the sticking time would increase by orders of magnitude if total angular momentum were not conserved. One may argue that this occurs in a static electric field. Unfortunately, however, the statistical theory cannot tell us at what strength of the electric field non-conservation of angular momentum occurs.

We present a quantitative theory of the required strength of such "symmetry-breaking interactions" based on classical simulations of collision complexes. We find that static electric fields as small as 10 V/cm can lead to non-conservation of angular momentum. Using the same framework, we also compute loss of collision complexes due to spontaneous emission and absorption of black-body radiation, which are found to be slow, and show that nuclear spin is conserved during sticky collisions.

Abstracts

Boerge Hemmerling

UC Riverside

"Target studies and spectroscopy for laser cooling of AlCl"

Ultracold polar molecules allow for a range of novel studies, including many-body physics of quantum degenerate gases, quantum computing, precision measurements and tests of fundamental symmetries. Laser cooling has been key as a first step towards implementing similar applications with atoms. Applying similar schemes to molecules is challenging due to their additional degrees of freedom, which interrupt the photon cycling process by a decay into dark states. With an estimated Franck-Condon factor of ~99.88%, AlCl is an excellent candidate for laser cooling. Starting with a cryogenic buffer-gas beam source, we use pulsed-laser ablation to produce AlCl in the gas phase and carry out precise spectroscopy on AlCl. We have systematically studied and compared various precursor targets, including mixtures of KCl:Al, NaCl:Al, $CaCl_2$:Al, $MgCl_2$:Al, and $AlCl_3$, and characterized their yield of AlCl, while monitoring other produced compounds. We will give an update on the status of the experiment, our target studies and discuss our progress towards slowing and cooling AlCl.

Eric Hudson

UCLA

"Optical Cycling of Molecular Ions and Quantum Functional Groups"

Compared to neutral molecules, molecular ions with optical cycling centers are surprisingly rare. We will summarize efforts to find molecular ions amenable to photon cycling and discuss high-resolution spectroscopy enabling use of one candidate, $^{29}SiO^+$, which appears to host a robust qubit. Finally, we will discuss recent work [1], and its implications, aimed at answering the question: is a universal quantum functional group possible? And if so, what can you do with it?

Nick Hutzler

Caltech

"Precision Measurements and New Directions with Polyatomic Molecules"

The extra degrees of freedom of polyatomic molecules allow the combination of multiple features which are incongruous in simpler systems, and enable design and tuning of molecules for desired applications. We will discuss experimental progress toward using polyatomic molecules for CP-violation searches, which benefit from the ability to combine photon cycling with high polarizability through parity doublets and the ability to control electromagnetic interactions. We will also discuss theoretical progress toward developing new motifs for engineering molecules with photon cycling centers with distinctly different properties than those previously developed, such as systems with multiple valence electrons and higher bond orders, which offer new opportunities and directions for fundamental and quantum science.