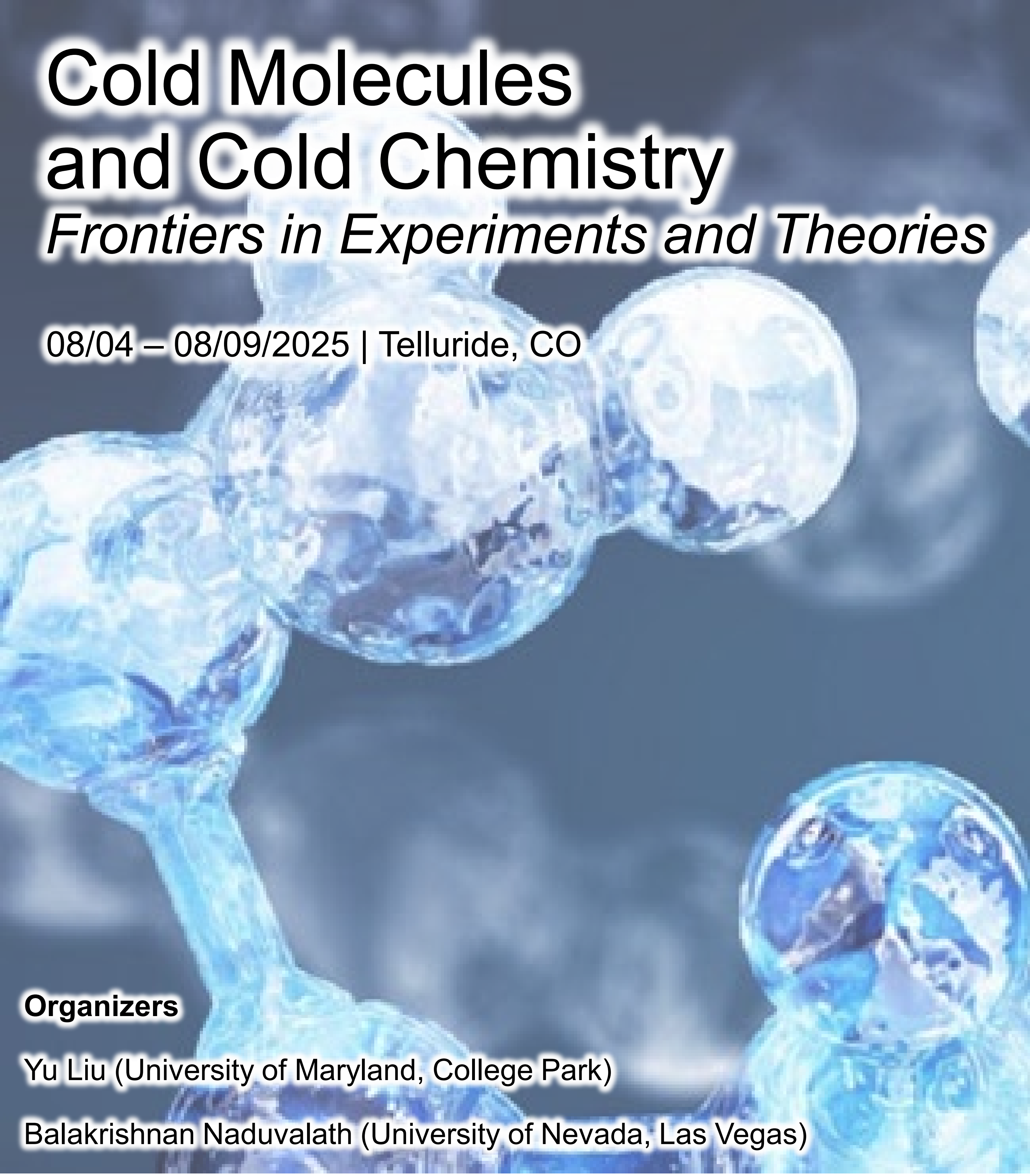

Cold Molecules and Cold Chemistry

Frontiers in Experiments and Theories

08/04 – 08/09/2025 | Telluride, CO

Organizers

Yu Liu (University of Maryland, College Park)

Balakrishnan Naduvalath (University of Nevada, Las Vegas)

TELLURIDE SCIENCE
& INNOVATION CENTER

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Day	Start Time	End Time	Event	Presenter	Title
Sunday (8/3)	17:00	18:30	Meet and greet at the Alibi (with cash bar)		
Monday (8/4)	7:30	8:30	Breakfast		
	8:45	9:00	Welcome remarks	Balakrishnan Naduvalath, Yu Liu	
	Session 1: Advances in direct laser cooling of molecules (session chair: Yuval Shagam)				
	9:00	9:50	Talk	Simon Scheidegger	Ultracold Yttrium Monoxide, Collisions and Narrowline Cooling
	9:50	10:40	Talk	Debayan Mitra	Directly cooled molecules for quantum chemistry and quantum simulations
	10:40	11:00	Coffee break		
	11:00	11:50	Talk	Ben Augenbraun (remote)	Hot Science with Cold Molecules: New Molecular Systems for Optical Cycling and Precision Measurements
	11:50	12:40	Talk	Haowen Zhou	Exploring chemistry at cold temperatures: From fundamental collision systems to potentially laser-coolable complex molecules
	12:40	14:30	Lunch break		
	Session 2: Advances in theory of cold collisions (session chair: Lee Liu)				
	14:30	15:20	Talk	Tijs Karman	Double microwave shielding
	15:20	16:10	Talk	Jesus Perez Rios	Buffer gas and ion chemistry: A theoretical study
	16:10	16:20	Coffee break		
	16:20	17:10	Talk	Balakrishnan Naduvalath	Stereodynamics of Cold Molecular Collisions
Tuesday (8/5)	7:30	8:30	Breakfast		
	Session 3: Ultracold collisions and reactions - I (session chair: Roman Krems)				
	8:45	9:35	Talk	Tiangang Yang (remote)	Precision-Controlled Cold Ion–Molecule Systems for Quantum-State-Resolved Reaction Dynamics
	9:35	10:25	Talk	Brian Kendrick	Non-adiabatic Quantum Dynamics of Ultracold Chemical Reactions
	10:25	10:45	Coffee break		
	10:45	11:35	Talk	Jeshurun Luke	Spin Dynamics and Quantum Coherence in Ultracold Molecular Reactions
	11:35	12:25	Talk	Timur V. Tscherbul	Ultracold short-range chemistry: Entanglement of collision products, hyperfine-to-rotational energy transfer, and low-scaling coupled-channel algorithms
	12:25	14:30	Lunch break		
	Session 4: Ultracold collisions and reactions - II (session chair: Bala Naduvalath)				
	14:30	15:20	Talk	Yu Liu	Precision Chemistry with Ultracold Molecules
	15:20	16:10	Talk	John Bohn	Applications of Ultracold Molecular Collisions
	16:10	16:30	Coffee break		
	16:30	17:20	Talk	Ed Narevicius	TBD
	17:20	18:10	Talk	Olivia Krohn	Novel method for ultrahigh velocity resolution in molecular beams for inelastic scattering dynamics and chemical reactions
	18:30	19:30	Telluride Town talk		
Wednesday (8/6)	7:30	8:30	Breakfast (grab and go)		

	8:30	2:00	Free for hiking or other activities		

Session 5: New theoretical tools for molecular structure and dynamics (Tijs Karman)

	13:30	14:20	Talk	Micheline Soley (remote)	Tensor Trains and Quantum Computing for Highly Multidimensional Molecular Simulations
	14:20	14:45	Talk	Jayakrushna Sahoo (contributed)	Simulating electronic nonadiabatic atom–molecule collisions on a quantum computer
	14:45	15:15	Coffee break		
	15:15	16:05	Talk	Roman Krems	How machine learning can accelerate quantum dynamics calculations
	16:05	16:55	Talk	Svetlana Kotochigova	Anisotropic Chemical Bonding of Lanthanide–OH Molecules
	17:30	19:30	Picnic		
Thursday (8/7)	7:30	8:30	Breakfast		

Session 6: Precision Spectroscopy of Cold Molecules (session chair: Yu Liu)

	8:45	9:35	Talk	Chin-Wen Chou	Direct-comb precision spectroscopy and quantum state control of single molecular ions
	9:35	10:25	Talk	Yuval Shagam	Searching for parity violation in trapped chiral molecular ions
	10:25	10:45	Coffee break		
	10:45	11:35	Talk	Michael Drewsen	Prospects of photon recoil spectroscopy of complex molecular ions
	11:35	12:25	Talk	Lee Liu	Shining new light on highly symmetric molecules
	12:25	-	Free for hiking or other activities		
Friday (8/8)	7:30	8:30	Breakfast		

Session 7: Advances in Quantum Control of Cold Molecules (session chair: Didi Leibfried)

	8:45	9:35	Talk	Baruch Margulis	Quantum state control of a rotationally cold molecular ion
	9:35	10:25	Talk	Ziv Meir	Coherent Control of a Molecular Isomer Superposition
	10:25	10:45	Coffee break		
	10:45	11:35	Talk	Stefan Willitsch	Quantum technologies for trapped molecular ions and their applications
	11:35		Concluding remarks	Balakrishnan Naduvalath, Yu Liu	

Session 1 (Monday Morning): Advances in direct laser cooling of molecules

Session chair: Yuval Shagam (Technion)

Ultracold Yttrium Monoxide, Collisions and Narrowline Cooling

Simon Scheidegger (JILA, NIST and University of Colorado, Boulder)

Co-authors: K. Mehling, J. J. Burau, M. Chen, L. E. Hillberry, Lan Cheng, Jun Ye (affiliations: JILA, NIST & University of Colorado Boulder; Johns Hopkins University)

Abstract: Laser cooling of paramagnetic diatomic molecules [1,2] is usually constrained by using a P(1) transition between an electronic $^2\Sigma^+$ ground state and an excited state with $^2\Sigma^+$ or $^2\Pi$ character. I will present our recent work on narrowline laser cooling using transitions between the X $^2\Sigma^+$ ground electronic state and the metastable A' $^2\Delta_{3/2}$ of yttrium monoxide. We identified a quasi-closed photon-cycling scheme addressing a single quantum state. By applying small electric fields we isolated the desired pure-parity state energetically from unwanted mixed-parity states. With this additional quantum control we regained the required photon-cycling closure and achieved the first experimental realization of narrowline laser cooling of a molecule [3]. I will further discuss the potential of this metastable $^2\Delta$ state for future experiments and report on our ongoing efforts to realize a collisionally stable gas of ultracold molecules in an optical dipole trap using the technique of microwave shielding.

[1] Ding et al., Physical Review X 10, 021049 (2020)

[2] J. J. Burau et al., Physical Review A 110, L041306 (2024)

[3] K. Mehling et al., arXiv 2503.13838

Directly cooled molecules for quantum chemistry and quantum simulations

Debayan Mitra (Indiana University Bloomington)

Abstract: In recent years, cold and ultracold molecules have emerged as a mature platform for quantum simulation, computation, chemistry, and precision measurements. Molecules provide unique features and challenges compared to their atomic analogs: they can strongly interact at long range due to their inherent dipole moments, and they offer internal states that are insensitive to external fields. Many molecular platforms have emerged in the last decade based on diatomic and polyatomic molecules. In this talk, I will discuss two avenues where molecular advantages play a key role. First, I will discuss how the molecule CaH can be used as a vehicle to produce ultracold, trapped hydrogen atoms for precision measurement. I will describe the formation of CaH and CaD molecules, how we model the process of chemical reaction in the cryogenic environment of a buffer-gas cell, and our latest efforts towards laser cooling CaH. Second, I will discuss a novel molecular quantum gas microscope designed around a fermionic diatomic molecule. A majority of emergent phenomena in the solid state arise from entanglement due to strong interactions between electrons and the interplay with the underlying lattice structure. I describe how the molecule MgF possesses many properties favorable to both laser cooling and single-site imaging. These molecules will interact via tunable dipole–dipole interactions while they quantum tunnel from site to site. With these tools, we can realize forms of itinerant quantum magnetism as well as strongly correlated quantum systems.

Hot Science with Cold Molecules: New Molecular Systems for Optical Cycling and Precision Measurements

Ben Augenbraun (Williams College)

Abstract: The broad scientific opportunities promised by ultracold molecules have spurred efforts to directly laser cool diverse classes of molecules. Increasing attention has been attracted by molecules that provide new resources for quantum science and precision measurements. This includes molecules with complex structure, where long-lived states present rich qubit platforms and/or internal co-magnetometers that are robust against external field fluctuations. In this talk, we introduce new potentially laser-coolable molecules: (1) coinage metals bonded to carbon-group atoms (e.g., AuC or AgPb) and (2) the lanthanide- containing molecule DyO. For distinct reasons, both of these groups of molecules possess easily polarizable ground states with negligible magnetic-field sensitivity, diagonal Franck–Condon factors that enable optical cycling and laser cooling, and relativistically enhanced sensitivity to physics beyond the Standard Model. In addition to presenting our group’s recent experimental progress studying AuC and DyO, we discuss the rational design of similar molecules that may be useful for future quantum science experiments.

Exploring chemistry at cold temperatures: From fundamental collision systems to potentially laser- coolable complex molecules

Haowen Zhou (University of California, Los Angeles)

Abstract: Chemistry at low temperatures allows us to reveal molecular interactions in unprecedented detail. Various approaches have been developed to bring more chemically relevant species into the cold and ultracold regime. In the first half of the talk, I present results of rotationally inelastic collisions between simple molecules such as H_2/D_2 and He/Ne at ~1 K. Performed in a single molecular beam, the cold collision between two co-expanded species yields clear partial-wave structures in the product angular distribution. However, discrepancies regarding the collision temperatures still remain. In the second half of the talk, I present attempts at bringing laser cooling capabilities into complex molecules. This approach relies on Ca/Sr as a laser-coolable optical cycling center, which is largely isolated from the rest of the molecule. Using this strategy, molecules with >30 atoms have been identified with favorable optical cycling properties. However, as the molecules grow in size, vibronic interactions among the high density of states can lead to critical failures and limit their use.

Session 2 (Monday Afternoon): Advances in theory of cold collisions

Session chair: Lee Liu (Purdue University)

Double microwave shielding

Tijs Karman (Radboud University, Nijmegen)

Abstract: I will present double microwave shielding, which has recently enabled evaporative cooling to the first Bose–Einstein condensate of polar molecules. Two microwave fields of different frequency and polarization are employed to effectively shield polar molecules from inelastic collisions and three-body recombination, thereby suppressing two- and three-body losses. Simultaneously, dipolar interactions and the scattering length can be flexibly tuned, enabling comprehensive control over interactions in ultracold gases of polar molecules. This approach works universally for a wide range of molecules, opening the door to studying many-body physics with strongly interacting dipolar quantum matter.

Buffer gas and ion chemistry: A theoretical study

Jesus Pérez-Ríos (Stony Brook University)

Abstract: Most molecules of interest in cold molecular science (except for bi-alkali) are synthesized in a buffer gas cell, where they are also cooled before being ready for laser cooling or MOT loading. A solid target is ablated in the presence of a He buffer gas, which cools the target molecules. However, it is more stable and reliable to synthesize a molecule via a chemical reaction, known as buffer-gas chemistry. In this talk, we present a theoretical approach to characterize the CaH yield after ablating Ca in the presence of hydrogen. The results, aligning well with experimental data, help identify unforeseen loss channels related to plasma shielding. Indeed, our methodology is extensible to other interesting scenarios for the efficient synthesis of cold molecules. On the other hand, a trapped ion at cryogenic temperatures is another interesting system in chemical physics for spectroscopy and the understanding of ion–neutral reactions. In that regard, we study ion–atom–atom recombination reactions using a classical trajectory method in hyperspherical coordinates, which agrees very well with experimental observations.

Stereodynamics of Cold Molecular Collisions

Balakrishnan Naduvalath (University of Nevada, Las Vegas)

Abstract: Advances in quantum state preparation combined with molecule cooling and trapping technologies have enabled unprecedented control of molecular collision dynamics. This progress, achieved over the last two decades, has dramatically improved our understanding of molecular phenomena in the extreme quantum regime characterized by translational temperatures well below a kelvin. In this regime, collision outcomes are dominated by isolated partial waves, threshold behavior, quantum statistical effects, tiny energy splittings at the spin and hyperfine levels, and long-range forces. Collision outcomes are influenced not only by the quantum state preparation of the initial molecular states but also by the polarization of their rotational angular momentum, i.e., the stereodynamics of molecular collisions. In this talk, I provide an overview of recent theoretical developments describing the stereodynamics of cold molecular collisions and discuss their implications for cold controlled chemistry.

Session 3 (Tuesday Morning): Ultracold collisions and reactions – I

Session chair: Roman Krems (University of British Colombia)

Precision-Controlled Cold Ion–Molecule Systems for Quantum-State-Resolved Reaction Dynamics

Tiangang Yang (Southern University of Science and Technology, China)

Abstract: Understanding chemical reactions at the quantum level requires precise control over molecular quantum states and collision energies, especially at low temperatures where quantum effects dominate. We present recent progress in developing integrated platforms for quantum-state-resolved studies of ion– molecule reaction dynamics. Our system combines a laser-cooled ion trap with high-resolution time-of- flight mass spectrometry and fluorescence imaging, enabling sub-kelvin cooling of Be+ and C+ ions with accurate micromotion compensation. We have also demonstrated two- dimensional cooling of Be+ ions without repump lasers, a technique that may extend the applicability of laser cooling to more complex systems. To prepare quantum-state-selected neutral reactants, we employ a stimulated Raman pumping system with a fast chopper for H_2 and N_2, and a cavity-enhanced IR excitation scheme that achieves >30% population in the CO (v = 3) overtone state. These tools enable us to investigate how vibrational excitation, orientation, and isotope effects influence reactivity under cold conditions. Together, these capabilities provide a robust platform for studying state-to-state dynamics in cold collisions and for benchmarking theoretical models of interaction potentials and non-adiabatic couplings. Recent results and future directions for quantum-controlled reactivity will be discussed .

Spin Dynamics and Quantum Coherence in Ultracold Molecular Reactions

Jeshurun Luke (Harvard University)

Abstract: Ultracold chemistry has enabled the study of chemical reactions under a new lens. Complete quantum state control has opened avenues towards observing various quantum phenomena, such as how the quantum statistics of reactants control reaction rates and accessible product channels. We examine the extent to which quantum coherence is preserved in the nuclear spin degree of freedom during the KRb + KRb molecule–molecule reaction. Our results indicate that nuclear spin coherence is maintained throughout the reaction, manifesting in destructive interference of product reaction channels. These findings suggest the possibility of redistributing entanglement from within a molecule to spatially separated reaction products. In the second half, we explore energy exchange between atomic hyperfine and molecular rotational degrees of freedom in an atomic Rb + KRb collision. The experimentally measured product-state distribution suggests a statistical outcome where spin and rotation are completely mixed. However, detailed quantum scattering calculations predict the conservation of total mechanical angular momentum. The experimental result, in contrast, indicates that additional factors – such as a submerged conical intersection and the involvement of vibrational degrees of freedom – may be necessary to fully describe the dynamics. These observations can serve as benchmarks for developing future theoretical models and may suggest a more active role for spins in chemical reactions.

Ultracold short-range chemistry: Entanglement of collision products, hyperfine-to-rotational energy transfer, and low-scaling coupled-channel algorithms

Timur V. Tscherbul (University of Nevada, Reno)

Abstract: Owing to recent experimental advances in the detection of reaction products, ultracold short-range chemistry now offers an exciting avenue towards probing microscopic interactions within few-body reaction complexes with unprecedented precision. In addition, ultracold chemical reactions provide a natural laboratory for creating entangled pairs of product molecules, which can potentially be used for quantum science applications. Exploring these opportunities requires converged coupled-channel calculations on the best available ab initio potential energy surfaces. However, such calculations are extremely time-consuming, due in part to the steep cubic scaling of current algorithms with the number of scattering channels. Here, we report progress toward the development of a quadratically-scaling iterative approach for calculating a single column of the S-matrix. The approach is based on the iterative solution of the multichannel Lippmann–Schwinger equation and is particularly well-suited for cold and ultracold collisions of molecules prepared in single quantum states. We extend the original Thomas algorithm to include closed channels as necessary to obtain converged scattering observables and apply it to cold He + CO collisions on an accurate ab initio potential energy surface. We observe a slowdown in convergence when closed channels are included, necessitating the use of additional convergence acceleration techniques, such as the Fredholm expansion.

Non-adiabatic Quantum Dynamics of Ultracold Chemical Reactions

Brian Kendrick (Los Alamos National Laboratory)

Abstract: Experimental techniques for cooling, trapping and colliding cold ($T < 1$ K) and ultracold ($T < 1$ mK) molecules continue to develop at an astounding pace. Recent techniques using external fields and/or initial quantum state selection have demonstrated exquisite control of collision outcomes, including chemical reactivity. Recent experiments have also identified long-lived collision complexes and measured their lifetimes – for example, in Rb + KRb collisions these lifetimes exceed statistical estimates by five orders of magnitude. Thus, an accurate theoretical treatment of ultracold chemical reactions is needed. However, accurate calculations are challenging and require a fully quantum mechanical, first-principles description of both the electronic and nuclear motion. A numerically exact quantum reactive scattering methodology for atom–diatom (A + BC) collisions/reactions will be reviewed. This methodology is ideally suited for treating ultracold chemical reactions and has recently identified a novel quantum interference mechanism that could be exploited to effectively turn a chemical reaction on or off (i.e., a quantum molecular switch). This mechanism is general and has been shown to occur in a variety of ultracold reactions (e.g., $O + OH \rightarrow H + O_2$; $H + HD \rightarrow H + HD$; $Li + LiNa \rightarrow Li_2 + Na$; $K + KRb \rightarrow K_2 + Rb$). Of particular interest is the crucial role of an excited electronic state that becomes degenerate with the ground electronic state. The geometric phase associated with this degeneracy is shown to control the constructive or destructive nature of the quantum interference and hence the

reaction outcome. For many ultracold reactions, the excited electronic state is energetically accessible even at ultracold collision temperatures. Thus, a fully coupled two-electronic-state (non-adiabatic) calculation is required.

Session 4 (Tuesday Afternoon): Ultracold collisions and reactions – II

Session chair: Balakrishnan Naduvalath (UNLV)

Precision Chemistry with Ultracold Molecules

Yu Liu (University of Maryland, College Park)

Co-authors: Jingchen Zhang, Paul Julienne (University of Maryland, College Park)

Abstract: Over the past few decades, concurrent advances in experimental techniques in both atomic, molecular, and optical (AMO) physics and physical chemistry have enabled unprecedented control over simple molecules, both in terms of their translational motion and their internal states. In this talk, I will discuss the potential for these fully-controlled molecules to advance our understanding and control of reaction dynamics at the quantum level. I will present a series of studies [1] on the reaction KRb + KRb → K_2 + Rb_2 initiated at $T < 1\ \mu K$ [2], including the direct observation of a long-lived reactive complex [3], the demonstration of product rotational state control via conserved nuclear spins [4], and a test of the statistical model using the pair-correlated quantum state distribution of the products [5]. These initial experiments paved the way for a new project in my group at the University of Maryland, where we are aiming to study isotope exchange reactions between Li atoms and diatomic molecules (e.g., ^{7}Li + ^{6}Li2 → ^{6}Li^{7}Li + ^{6}Li, ^{6}Li7Li + ^{6}Li7Li → ^{6}Li2 + ^{7}Li^{2}) at ultralow temperatures, and at the state-to-state level. I will present the prospect of this system as a playground for investigating quantum effects in reaction dynamics, such as interference between reaction pathways, entanglement between reaction products, and coherent control of bimolecular reactions. Finally, I will present our recent theory work on possible pathways to generate the ultracold reactants, specifically the formation of ultracold Li_2 dimers from laser cooled Li atoms.

[1] Liu, Y., & Ni, K.-K. Bimolecular Chemistry in the Ultracold Regime. *Annual Review of Physical Chemistry*, 73(1) (2022).

[2] Hu, M. G., Liu, Y., … Ni, K. K. Direct observation of bimolecular reactions of ultracold KRb molecules. *Science* (New York, N.Y.), 366(6469), 1111–1115 (2019).

[3] Liu, Y., Hu, M. G., … Ni, K. K. Photo-excitation of long-lived transient intermediates in ultracold reactions. *Nature Physics* (2020).

[4] Hu, M. G., Liu, Y., … Ni, K. K. Nuclear spin conservation enables state-to-state control of ultracold molecular reactions. *Nature Chemistry* (2020).

[5] Liu, Y., Hu, M.-G., Nichols, M. A., Yang, D., Xie, D., Guo, H., & Ni, K.-K. Precision test of statistical dynamics with state-to-state ultracold chemistry. *Nature*, 593(7859), 379–384 (2021).

Applications of Ultracold Molecular Collisions

John L. Bohn (JILA, University of Colorado Boulder)

Abstract: Molecular collisions in the ultracold regime are often well-understood in terms of the purely long- range forces between the collision partners. In this circumstance, the usual complexities of determining potential energy surfaces and carrying out large-scale scattering calculations are greatly simplified. Because of this, in this talk I will not dwell on the calculation of the scattering itself, but rather on the applications of scattering results to situations relevant to ultracold systems. These include evaporative cooling, molecular entanglement, and the hydrodynamic behavior of molecular gases.

Title TBD

Edvardas Narevicius (TU Dortmund)

Abstract: TBD

Novel method for ultrahigh velocity resolution in molecular beams for inelastic scattering dynamics and chemical reactions

Olivia Krohn (Sandia National Laboratory)

Abstract: The study of inelastic collisions in the gas-phase remains an excellent probe of the fundamental potential energy surface of the interacting molecules. Such studies are often involve measuring the differential cross section of the scattered products as a function of energy and quantum state of the final product. These studies are typically conducted within molecular beams, which provide rotationally cold molecules, and with velocity map ion imaging (VMII), which enables state-selective and velocity sensitive detection of one the final scattering product states. These tools have been prolific in studies of atomic-molecular systems; however, the detection resolution of both final products states in bi-molecular collisions has been historically impeded by the velocity width of the molecular beam, which blurs the kinetic energy resolution of the products. For this reason, an approachable method to achieve excellent velocity resolution is extremely impactful to studies of molecular collisions. We present a technique to utilize co-linear mid-infrared (MIR) and molecular beams to prepare molecules in a chosen ro-vibrational quantum state with extremely high velocity resolution. The MIR laser counter-

propagates to the molecular beam, such that the center wavelength and linewidth dictate the center velocity and velocity width, respectively, of the excited molecules. With the achieved velocity resolution[1], studies of bi-molecular collisions with full information of both final products will be achievable. I will discuss the capabilities of this technique, as well as present current and planned studies of inelastic scattering of nitric oxide.

[1] O. A. Krohn and David W. Chandler. The Journal of Physical Chemistry Letters 2024 15 (50), 12455-12463. DOI: 10.1021/acs.jpclett.4c02927

Session 5 (Wednesday Afternoon): New theoretical tools for molecular structure and dynamics

Session chair: Tijs Karman (Nijmegen)

Tensor Trains and Quantum Computing for Highly Multidimensional Molecular Simulations

Micheline Soley (University of Wisconsin–Madison)

Abstract: Today, simulation of ultracold chemical reactions with exact quantum dynamics faces the “curse of dimensionality,” in which computational cost grows exponentially as the dimensionality of quantum systems increases. This limits exact grid-based quantum dynamics simulations to the smallest systems. Low-rank tensor-network approaches provide a way to surmount this curse for many quantum systems, as they can exponentially reduce computational cost for weakly coupled systems. Our work capitalizes on the native advantages of tensor networks and the high degree of entanglement possible on quantum computers to develop new approaches to simulate quantum systems in high dimensionality. In addition, we demonstrate that standard ultracold-atom scattering experiments can be used to reveal the existence of long-sought-after quantum mechanical parity-time reversal (PT) symmetry behavior using reflectionless scattering modes – a discovery which opens the door to the development of powerful quantum technologies.

Simulating Electronic Nonadiabatic Atom–Molecule Collisions on a Quantum Computer

Jayakrushna Sahoo (New Mexico Consortium, Los Alamos, NM)

Co-authors: Jonathan Andrade-Plascencia, Tamila Kuanysheva, Brian K. Kendrick, Dmitri Babikov (affiliations: Marquette University; Cornell University; Los Alamos National Laboratory; Marquette University)

Abstract: We propose a quantum algorithm to study electronic nonadiabatic inelastic collisions between an atom and a molecule, formulated within the framework of an Ehrenfest-type mixed quantum/classical dynamics. In this approach, nuclear motion is treated classically via Hamilton's equations, while electronic dynamics are described quantum mechanically using the time-dependent Schrödinger equation. A symmetrized second-order Suzuki–Trotter decomposition is employed to implement the time evolution of the electronic amplitudes on a quantum computer. The electronic Hamiltonian matrix is mapped to a qubit basis and expressed as a linear combination of Pauli operators. As a proof-of-principle demonstration, the algorithm is applied to the Ar + NO collision system. The quantum algorithm is tested both on a classical emulator and on actual quantum hardware, under the so-called adiabatic-trajectory approximation, and excellent agreement is observed between the results of the quantum and classical simulations. Future directions include incorporating a quantum mechanical treatment of molecular rotation into the electronic Hamiltonian.

How machine learning can accelerate quantum dynamics calculations

Roman V. Krems (University of British Columbia)

Abstract: Supervised machine learning is often used as an efficient interpolation tool in multi-dimensional spaces. With some effort, machine learning models can be designed to extrapolate. I discuss extrapolation by machine learning for the purpose of reducing the numerical difficulty of quantum dynamics calculations. In particular, I show that Bayesian models can be designed to extrapolate observables from Hilbert spaces of lower dimensionality to predict observables that require larger basis sets to compute. I describe a general approach that combines machine learning and numerical integration of the Schrödinger equation into a unified framework that can be used for (1) solving inverse quantum problems by Bayesian optimization; (2) identifying the sensitivity of quantum observables to details of the underlying potential energy surfaces and determining error bars of dynamical calculations stemming from uncertainties of quantum chemistry calculations; (3) accelerating quantum dynamics calculations by extrapolation in Hamiltonian parameter spaces. Time permitting, I would also like to discuss a bridge between molecular dynamics and quantum computing through quantum machine learning (defined as machine learning with part of the algorithm implemented on a quantum computer).

Anisotropic Chemical Bonding of Lanthanide–OH Molecules

Svetlana Kotochigova (Temple University)

Abstract: We describe calculations of the relativistic electronic states of magnetic DyOH and ErOH using restricted-active-space self-consistent-field methods combined with computed spin–orbit matrix elements, as well as a relativistic coupled-cluster method. With the self-consistent-field method, we also compute electric and magnetic dipole moments. We find that the equilibrium geometry is linear with the oxygen atom in the middle, and that the magnetic lanthanide atom loses one electron to OH. Most importantly, this electron originates from the submerged open 4f shell of the lanthanide atom and mixes with the $2p^5$ configuration of the tightly bound ground-state OH. The "spin–orbit" levels of the 4f^(n– 1)6s^2 + 2p^6 configuration (with n = 10 for DyOH and n = 12 for ErOH) correspond to the ground and lowest-energy excited states. The bond is distinctly different from that predicted by density functional theory (DFT), in which the lanthanide atom loses one electron from its outer 6s shell to OH. Other groups found a similar bond in the isoelectronic DyF molecule using four-component relativistic configuration- interaction approaches.

Session 6 (Thursday Morning): Precision Spectroscopy of Cold Molecules

Session chair: Yu Liu (University of Maryland, College Park)

Direct-comb precision spectroscopy and quantum state control of single molecular ions

Chin-wen Chou (NIST, Boulder & University of Colorado, Boulder)

Co-authors: Zhimin Liu, Yu Liu, Joaquin Berrocal Sánchez, April Sheffield, Peter Chang, Sichong Ma, Tsung- Han Wu, April Reisenfeld, Dietrich Leibfried, Scott Diddams (affiliations: NIST, Boulder, CO; University of Colorado, Boulder, CO; University of Colorado, Denver, CO; University of Maryland, College Park, MD)

Abstract: Quantum state control of cold molecules promises advances in disciplines including precision measurements, quantum information processing, and chemical physics. Equipped with single-molecular quantum-state manipulations enabled by quantum-logic spectroscopy, we perform precision rotational and vibrational spectroscopy of a single CaH+ ion. Starting with the molecule prepared in a pure quantum state, frequency-comb light is directly employed to coherently excite rotational and vibrational transitions via single-photon or two-photon Raman processes. The transition frequencies can be determined with better than one-part-per-trillion precision. Furthermore, we are developing a dual-branch frequency comb system aiming for agile driving and precision spectroscopy of multiple vibrational transitions in the ground electronic state of CaH+. The frequency-comb-based light sources could enable coherent preparation and analysis of molecular vibrational wavepackets and general

coherent superpositions of vibrational states. Improved and expanded characterization of molecular properties and state control in turn opens up possibilities for applications of molecules in quantum sciences.

Searching for parity violation in trapped chiral molecular ions

Yuval Shagam (Technion – Israel Institute of Technology)

Abstract: The weak force is predicted to break the parity symmetry between left- and right-handed chiral molecules, but so far the effect has eluded detection. We are developing a trapped chiral molecular ion version of the search for parity violation in molecules. Our candidate molecule, CHDBrI+, is predicted to be preparable via state-selective ionization and to exhibit a large parity-violating frequency shift of a few Hz for a C–H bend vibrational transition, where the transition's natural linewidth is narrower than the shift. Other transitions, such as the C–H stretch, are also predicted to have Hz-scale PV shifts between enantiomers. We plan to probe the PV signature in a racemic, mixed-handedness ensemble of trapped CHDBrI+ using vibrational Ramsey spectroscopy. We will discuss our experimental progress toward preparing cold chiral molecular ions by photoionization in the VUV regime. We will also discuss the advantages that chiral molecules have in searches for new physics, as well as the current status of the experiment.

Prospects of photon recoil spectroscopy of complex molecular ions

Michael Drewsen (Aarhus University)

Abstract: Trapped, single complex molecular ions have in the past been demonstrated to be sympathetically cooled translationally to near the Doppler laser cooling limit through the Coulomb interaction with laser cooled and co-trapped single atomic ions [1]. Furthermore, simpler molecular ions have been sympathetically cooled of to near their motional ground state through resolved sideband cooling of the atomic ions [2,3], and their internal state have been prepared either by buffer gas cooling [4] or probabilistic state detection [5-7]. In the talk, I will discuss, our efforts to extend ground state cooling of complex molecular ions with photon recoil spectroscopy in both the sideband unresolved [8] and resolved regime [9] with the aim - among others - of measuring the chirality of single molecules through a photon recoil spectroscopy version of the schemes presented in in Ref. [10]. Finally, to illustrate the very diverse potential of recoil spectroscopy, an example of how to test for potential extensions to quantum mechanics with charged macromolecules [11] will briefly be discussed.

[1] K. Højbjerre et al., Phys. Rev. A 77, 030702(R) (2008).

[2] G. Poulsen, PhD thesis, Aarhus University, 2011.

[3] R. Rugango et al., New J. Phys. 17, 035009 (2015).

[4] K. Hansen et al., Nature 508, 76 (2014).

[5] Wolf, F. et al., Nature 530, 457 (2016).

[6] C.-W. Chou et al., Nature 545, 203 (2017).

[7] M. Sinhal et al., Science 367, 1213 (2020).

[8] E. H. Clausen et al., Phys. Rev. A 105, 063709 (2022).

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Shining new light on highly symmetric molecules

Lee Liu (Purdue University)

Abstract: The C_{60} fullerene molecule has been the subject of intense study for four decades, starting with its identification in mass spectra of carbon soot in 1985. Following the observation of quantum-state-resolved infrared spectra in 2019, C_{60} is now the largest and most symmetric molecule for which rovibrational quantum state resolution has been achieved. We discuss recent results on laboratory spectroscopy of gas-phase C_{60}, including investigations of its dynamics, collision properties, and nuclear spin statistics. This motivates quantum state-resolved spectroscopy of large symmetric molecules as a window into the properties of complex quantum systems with symmetries and degrees of freedom not readily accessible in other composite systems. We also address the outstanding problem of restrictive selection rules in probing highly symmetric molecules.

Session 7 (Friday Morning): Advances in Quantum Control of Cold Molecular Ions

Session chair: Dietrich Leibfried (NIST)

Quantum state control of a rotationally cold molecular ion

Baruch Margulis (National Institute of Standards and Technology, Boulder)

Abstract: Over the last decade, molecules have emerged as a platform for endeavors such as the search for new physics, quantum information and simulation, and control of chemical reactions. However, unlike atoms, molecules vibrate and rotate, which makes them harder to cool and control at the single quantum- state level. For a trapped polar molecular ion such as ^40CaH+, the extent of the populated state space is affected by the blackbody radiation induced by the environment, resulting in a vast number of occupied states in a room-temperature trap and a significant challenge in quantum state control. In this talk I will present results from a new experiment, where a molecular ion is trapped in a cryogenic environment. We use quantum-logic spectroscopy on a ^40Ca+–^40CaH+ system to interrogate the molecular spin- rotational states. Operating in a cryogenic environment concentrates the rotational population to around 97% in J = 1–3 and results in rotational lifetimes of ~18 s for J = 1 (and ~10 s for J = 2). Utilizing the low rotational transition rates, we were able to demonstrate molecular state preparation and measurement (SPAM) fidelity of 99.8%. Additionally, I will describe our efforts towards loading other molecular species such as N_2+ and CO+ using a molecular beam combined with photoionization by a pulsed dye laser.

Coherent Control of a Molecular Isomer Superposition

Ziv Meir (Weizmann Institute of Science, Israel)

Co-author: Tamar Levin (Weizmann Institute of Science)

Abstract: Preserving quantum coherence becomes increasingly challenging as systems grow in size and complexity. Molecules, with their rich internal structure, offer a unique playground for studying the transition from quantum to classical behavior. Here, we present a scheme to coherently couple two distinct molecular isomers of the same molecule – specifically, the I = 0 and I = 2 nuclear-spin isomers of the nitrogen molecular ion. Our approach leverages an avoided crossing in the nitrogen-ion spectrum that mixes the two isomers via an electric-quadrupole hyperfine interaction. We show how to use this avoided crossing to strongly and coherently couple two unmixed nuclear-spin-isomer states, thereby creating a nuclear-spin-isomer qubit.

Quantum technologies for trapped molecular ions and their applications

Stefan Willitsch (University of Basel)

Abstract: Molecules are quantum systems of prime significance in a variety of contexts ranging from physics and chemistry to biology. In spite of their importance, the

development of quantum technologies for molecules has remained a long-standing challenge due to their complex energy-level structures. Trapped molecular ions are particularly attractive in this context, as it is possible to observe, manipulate and control single isolated molecules under precisely controlled conditions. In this talk, we highlight new experimental methods for the detection, preparation and manipulation of the quantum states of single trapped molecular ions and discuss applications of these techniques in the realms of precision molecular spectroscopy, quantum science and chemistry.