Paul Scheier, Anna Maria Reider, Elisabeth Gruber (Eds.)

16th Conference on Quantum Fluid Clusters 2026 (QFC 2026)

May 31st – June 4th, 2026
Obergurgl, Austria

Contributions

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Paul Scheier, Anna Maria Reider, Elisabeth Gruber

Preface

The QFC 2026 is the 16th occurrence of the QFC conference series. The focus of this interdisciplinary conference is to stimulate the research on atomic clusters, in particular helium nanodroplets, which are strongly influenced in their properties by quantum mechanical many-body effects. It covers both fundamental aspects such as superfluidity and quantum vortices, as well as application of QFC as flying nano-cryostats for probing and synthesizing cold molecules and exotic nano-complexes. The conference gathers theoreticians and experimentalists from the traditional fields of low-temperature physics, cluster physics, molecular spectroscopy, mass spectrometry, cold chemistry, laboratory astrophysics, ultrafast spectroscopy, nanoparticle imaging, etc.

Program

	Sunday May 31	Monday June 1	Tuesday June 2	Wednesday June 3	Thursday June 4
07:00 – 09:00	Breakfast	Breakfast	Breakfast	Breakfast	Breakfast
08:45 – 09:00		Opening			Departure
09:00 – 09:40	COSY COST Action – WG4 meeting	Valery Milner	Frank Stienkemeier	Alessandro Colombo	
09:40 – 10:20		Wenbin Zhang	Sergiy Krasnokutskiy	Wei Kong	
10:20 – 10:45		Mia Aomura	Florian Foitzik	Jeppe Kjædegaard	
10:45 – 11:15		Coffee	Coffee	Coffee	
11:15 – 11:55		Henrik Stapelfeldt	Devendra Mani	Marcel Mudrich	
11:55 – 12:35		Florent Calvo	Daniel Thomas	Jan-Michael Rost	
12:40 – 14:00		Lunch	Lunch	Lunch	
14:00 – 14:40		Susumu Kuma	Free Time	Javier Hernández Rojas	
14:40 – 15:20		Tomás González Lezana		Marta Hernández	
15:20 – 15:45		Francesco Ancilotto		Emil Hansen	
15:45 – 16:15		Coffee	Coffee	Coffee	
16:15 – 16:55	Registration for QFC	Nadine Halberstadt	Markus Koch	Christian Medina	16:15 – 16:40
16:55 – 17:35		Lukas Bruder	Shengfu Yang	Sivarama Krishnan	16:40 – 17:05
17:35 – 18:00		Matthias Vetenik	Juraj Fedor	Andrey Vilesov	17:05 – 17:45
				Closing	17:45 – 18:00
18:00 – 20:00	Dinner	Dinner	Dinner	Dinner	
20:00 – 22:00		Board Meeting	Poster Session		

Invited talk (30+10min)

Hot topic talk (20+5min)

Contents

Invited Talks

Control of molecular rotation in helium nanodroplets with an optical centrifuge

Valery Milner[1], Ian MacPhail-Bartley[1], Sören Mahr[1], Cameron Peters[1], Frank Stienkemeier[2], Henrik Stapelfeldt[3]

[1]*Department of Physics & Astronomy, University of British Columbia, Vancouver, Canada*

[2]*Institute of Physics, University of Freiburg, Freiburg, Germany*

[3]*Department of Chemistry, Aarhus University, Aarhus, Denmark*

We experimentally demonstrate that the rotation of molecules embedded in helium nanodroplets can be controlled with an optical centrifuge, allowing for the direct observation and study of molecular dynamics inside the strongly interacting many-body environment of superfluid helium.

I will discuss two new techniques to spin molecules inside helium nanodroplets. Fist, I will show that a laser pulse, whose polarization is rotating at a constant angular rate – a so-called "constant-frequency centrifuge", is capable of forcing molecules to follow the rotation of the optical field. When the frequency is resonant with a rotational transition, it is shown to persist on a nanosecond time scale, representative of superfluid environment.

I will then present our latest results on spinning up molecules with an "ultra-slow centrifuge", whose rotational frequency is ramping up with an extremely slow acceleration. Here, we observe different ways in which different molecular species are following the accelerated rotation, which may reveal important aspects about the interaction mechanisms between the molecule and the superfluid bath.

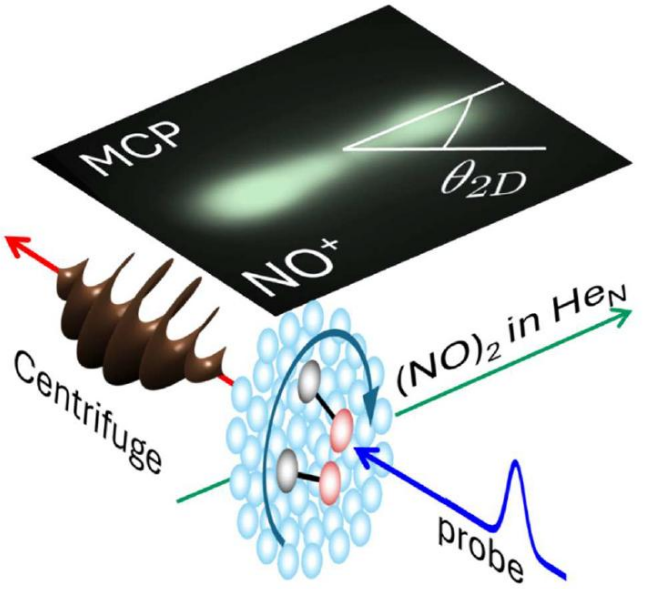

Figure 1: Illustration of the experimental approach: $(NO)_2$ spun up by a "constant-frequency optical centrifuge"

Delocalization and localization of small molecules in 0.37 K helium nanodroplets

Zhengjun Ye[1], Jiaxuan Chen[1], Haomai Hou[1], Junjie Qiang[1], Lianrong Zhou[1], Hongcheng Ni[1], Wenbin Zhang[1]* and Jian Wu[1]

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Superfluid helium nanodroplets act as ideal nanoreactors, cooling embedded molecules to 0.37 K and facilitating unique light-matter interactions. By using helium-nanodroplet target recoil ion momentum spectroscopy (He_NTRIMS) setup, we have intensively studied the ultrafast dynamics of these cold, in-droplet molecules [1-4]. Previous femtosecond pump-probe measurements from our group revealed diverse dynamics: while neutral D_2 rotates quasi-freely for >100 ps, charged ions (D_2^+) experience rapid collision-induced decoherence (~ 140 fs). Furthermore, strong ion-helium interactions drastically alter fragmentation dynamics of H_2 molecules.

Building on these interaction dynamics, we explore the spatial extent of a molecule's wave function with respect to a He_N by examining photoelectron momentum distributions (PMDs) from above-threshold ionization [5]. For the lightest H_2 molecule at 0.37 K, the thermal de Broglie wave expands significantly. Because this spatial extent approaches the droplet size, the wave function delocalizes across the droplet volume. This delocalization minimizes electron scattering with the helium surroundings, preserving distinct angular nodal structures in the PMDs. Conversely, heavier D_2 molecules possess narrower wave functions and localize deeply within the potential well. Photoelectrons emitted from these localized species undergo multiple undirected scatterings with the helium environment, resulting in heavily blurred nodal structures. Finally, we demonstrate how these contrasting quantum localization and delocalization behaviors significantly shape the ultrafast formation

dynamics of H_3^+ and D_3^+ from dimers confined in helium nanodroplets via engineered nuclear quantum effects [6].

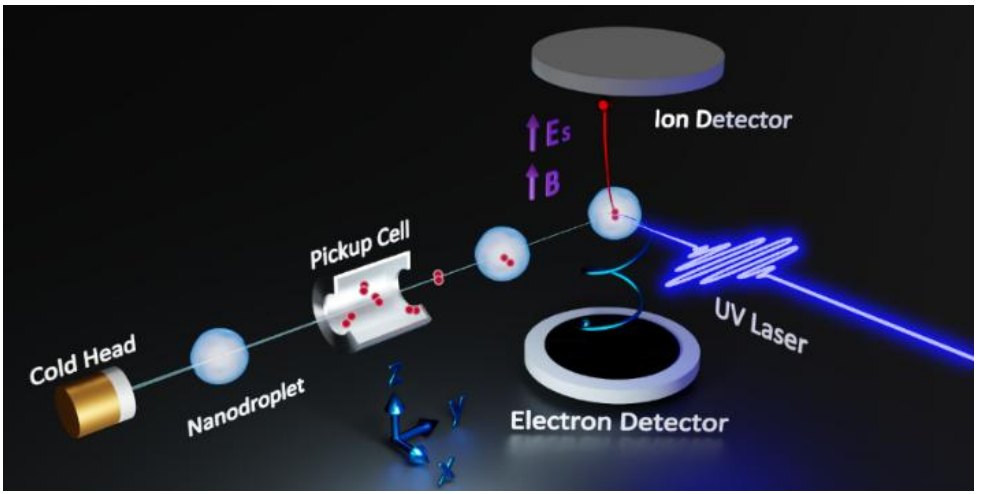

Figure 2: The experimental Setup

References:
1. J. Qiang et al, *Phys. Rev. Lett.* (2022), 128, 243201.
2. J. Qiang et al, *Phys. Rev. Lett.* (2024), 132, 103201.
3. L. Zhou et al, Phys. *Rev. Lett.* (2023), 130, 033201.
4. Z. Ye et al., Phys. *Rev. A.* (2025), 112, 063118.
5. Z. Ye et al., Phys. *Rev. Lett.* (2025), 135, 213202.
6. J. Chen et al., *in revision* (2026).

Real time observation of the diffusion-limited formation of an ion-molecule complex

Henrik Stapelfeldt, Jeppe K. Christensen
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Time-resolved studies of bond making between two molecules or atoms are notoriously difficult due to the challenge of measuring when the two reactants meet. This is determined by diffusion – a process that is typically not controllable on ultrafast time scales. Previous works used a weakly-bonded precursor complex of the two reactants to eliminate diffusion or introduced one of the reactants in a dense environment of reactants to minimize diffusion or allow it to be simulated by models.

I will present real-time measurements of the bimolecular reaction where a Li^+ ion diffuses towards a benzene dimer and forms a Li^+Bz_2 complex, a textbook cation-π-system, inside a liquid helium nanodroplet [1]. After formation at the droplet surface, Li^+ solvates [2-3], then diffuses ballistically with a velocity of 43 m/s and finally binds to Bz_2 near the droplet center to form Li^+Bz_2.
The experimental findings are compared to and rationalized by ring-polymer molecular dynamics simulations. Results from ongoing studies of real-time imaging of stereodynamics, i.e. how the molecular orientation changes as the ion approaches, will also be presented.

References:
1. J. K. Christensen *et al., Real-time observation of the diffusion-limited formation of a cation-molecule complex. Nature* Comm. **17**, 1249 (2025).
2. S. H. Albrechtsen *et al.*, Observing the primary steps of ion solvation in helium droplets. *Nature* **623**, 319 (2023).
3. J. K. Christensen *et al., Time-resolved solvation dynamics of Li+, Na+ and K+ ions in liquid helium nanodroplets.* Phys. Chem. Chem. Phys., **27**, 24184 (2025).

Optimal core-shell clusters of para-H_2/ortho-D_2

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Hydrogen and deuterium mix as solid solutions in the bulk. Recently, experimental evidence using Raman spectroscopy was provided in support of phase separation of H_2/D_2 clusters into core-shell structures, assisted by helium as a (super)cooling medium. In this contribution we explore the role of helium on the formation of core-shell structures by means of path-integral molecular dynamics simulations. More generally, the combined roles of size and composition are explored using harmonic vibrational analyses. While the core-shell pattern with deuterium in the core generally leads to the lowest zero-point energy, hence the greatest stability, other types of phase separation such as reverse core-shell or Janus-like are examined. We find that for any given composition there is an optimal core-shell cluster size minimizing the zero-point energy, and vice-versa. Our methodology also explains why such results cannot extrapolate to the bulk, the solid solution being almost isoenergetic to the randomly mixed phase.

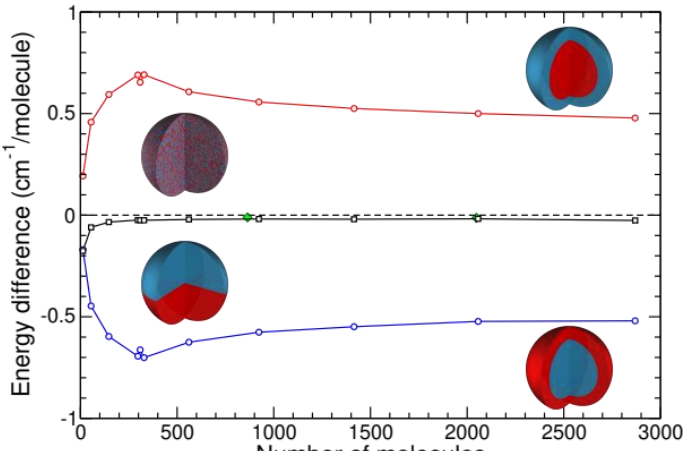

Figure 3: Energy of core-shell, reverse core-shell and Janus-like icosahedral $(H_2)_n(D_2)_n$ clusters relative to randomly mixed clusters, as a function of their size n.

References:

1. R. Sliter, K. Hyeon-Deuk, and A. F. Vilesov, *Phys. Rev. Lett.* **132**, 206001 (2024).

Water and Ammonia Radical Cluster Cations in Helium Droplets

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Helium nanodroplets serve as an ideal host for spectroscopic investigations of cations and reactive intermediates. Helium droplets are homogeneous and exist in the superfluid state, characterized by a low temperature of 0.4 K. Previously, a variety of cations produced by electron impact ionization of doped droplets were investigated using spectroscopy in the infrared spectral range. It was observed that electron impact ionization of helium nanodroplets doped with several protic molecules, such as H_2O and NH_3, yields not only protonated clusters (e.g., $(H_2O)_nH^+$) but also radical-cation clusters, such as $(H_2O)_n^+$. Radical-cation clusters are a significant class of reaction intermediates in radiation chemistry. With this technique, we successfully observed the hemibonded form of the water dimer cation $[H_2O–OH_2]^+$ for the first time [1], which competes with the more stable proton-transferred isomer (H_3O^+)–OH. While spectroscopic studies of protonated clusters in molecular beams are extensively reported, radical-cation clusters remain relatively unexplored due to the low yield of such clusters in conventional molecular beam experiments.

As an extension of the above study based on the method developed by the Vilesov group [2], we further investigated the ammonia dimer cations $(NH_3)_2^+$ and the larger cluster cations $(NH_3)_n^+$ and $(H_2O)_n^+$ (n = 3–6). In the experiments, we prepared corresponding neutral clusters in helium droplets prior to ionization while controlling the vapor pressure of water and ammonia. The vibrational spectra in the O–H and N–H stretching regions were subsequently measured using
a nanosecond pulsed mid-IR OPO laser in the 3–μm spectral region. The obtained vibrational peaks were assigned to the isomers by comparing them with the gas-phase and Ar-tagged spectra, as well as with the results of quantum chemistry calculations. Notably, the hemibonded isomer of

the ammonia dimer cations $[H_3N–NH_3]^+$ was not observed, which indicates the necessity of consideration for the trajectories on the potential energy surfaces during ionization in the presence of the He environment [3]. The spectral analysis of larger cluster cations of ammonia [4] and water [5] further elucidates the information on the ionized hydrogen-bonding network. Additionally, the water–ammonia binary cations is a good example of the study of ion-molecule reactions proceeding in helium droplets [6].

References:

1. A. Iguchi, A. Singh, S. Bergmeister, A. A. Azhagesan, K. Mizuse, A. Fujii, H. Tanuma, T. Azuma, P. Scheier, S. Kuma, and A. F. Vilesov, *J. Phys. Chem. Lett.* **14**, 8199 (2023).
2. D. Verma, S. Erukala, and A. F. Vilesov, *J. Phys. Chem. A* **124**, 6207 (2020).
3. A. Singh, A. Iguchi, T. C. Bernaards, Z. Golpariani, K. Mizuse, A. Fujii, H. Tanuma, T. Azuma, S. Kuma, and A. Vilesov, *J. Phys. Chem. A* **129**, 2472 (2025).
4. A. Singh, A. Iguchi, Z. Golpariani, T. C. Bernaards, A. Fujii, H. Tanuma, T. Azuma, K. Mizuse, S. Kuma, and A. F. Vilesov, *J. Phys. Chem. A* **129**, 9721 (2025).
5. A. Singh, A. Iguchi, Z. Golpariani, T. C. Bernaards, A. Fujii, H. Tanuma, T. Azuma, K. Mizuse, S. Kuma, and A. F. Vilesov, in preparation.
6. T. C. Bernaards, A. Singh, Z. Golpariani, A. Iguchi, P. Nehrudass, S. Kuma, and A. F. Vilesov, *J. Phys. Chem. Lett.* **17**, 804 (2026).

Solvation of ionic species in quantum fluid clusters: From Mozartkugel-like structures to spectroscopically relevant effects

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In the past recent years, we have been theoretically investigating the formation of large ordered structures in doped quantum clusters in close collaboration with the experiment. Multiply charged cations such as Ca^{2+} [1], Ho^{2+} [2] or Bi^{2+} [2] have been found to be effectively solvated in helium by means of concentric symmetric solvation shells involving up to 90 atoms. Attempts to link the occurrence of specific geometries in each case with the interatomic potentials describing the inside interactions have been suggested. In this communication we will try to review our current knowledge on this issue. Finally, we will report how previous studies on solvated polycyclic aromatic hydrocarbons such as coronene [3,4] have been extended to understand the spectroscopical investigation of Mg-decorated phthalocyanine.

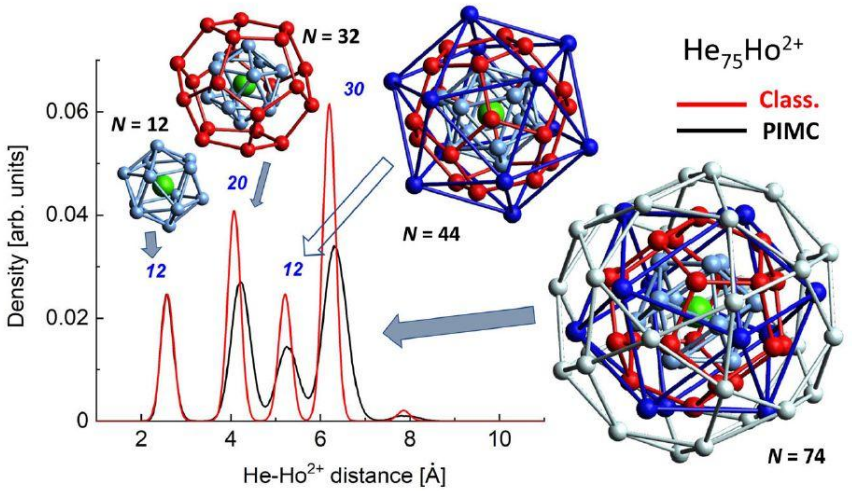

Figure 4: Probability density for interparticle distances and solvation shell structures for He_nHo^{2+} clusters.

References:

1. E. Zunzunegui-Bru *et al., J. Phys. Chem. Lett.* (2023), **14**, 3126-3131.
2. F. Foitzik *et al., Small Struct.* (2025), **6**, 2500094.
3. R. Rodríguez-Cantano *et al., J. Chem. Phys.* (2015), **143**, 224306.
4. R. Rodríguez-Cantano *et al., J. Chem. Phys.* (2017), **146**, 034302.

Real Time Dynamics Simulation of Pure and Doped Helium Nanodroplets

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[3]Institute of Nanoscience and Nanotechnology (IN2UB), Universitat de Barcelona, Spain; and Departament FQA, Facultat de Física, Universitat de Barcelona, Spain

The time-dependent dynamics of doped or pure helium nanodroplets poses numerous challenges to theoreticians, due to the highly quantum nature of this unusual ``solvent'' and to its eminent superfluid properties. After a short overview of existing theoretical methods, the Helium Time-dependent Density Functional Theory (4He-TDDFT) will be introduced. This method, describing the time evolution of the helium density rather than that of the many-body wave function, results from a compromise between accuracy and feasibility. It has emerged as a powerful tool to simulate and help understand many experimental results over the years.[1]

The rest of this Introductory talk will focus on some of the dynamical processes recently studied with this method in our group:

- A direct view on time-dependent ion solvation[2,3] (see illustration in Fig. 1)
- Coulomb explosion of dialkali molecules on the droplet surface

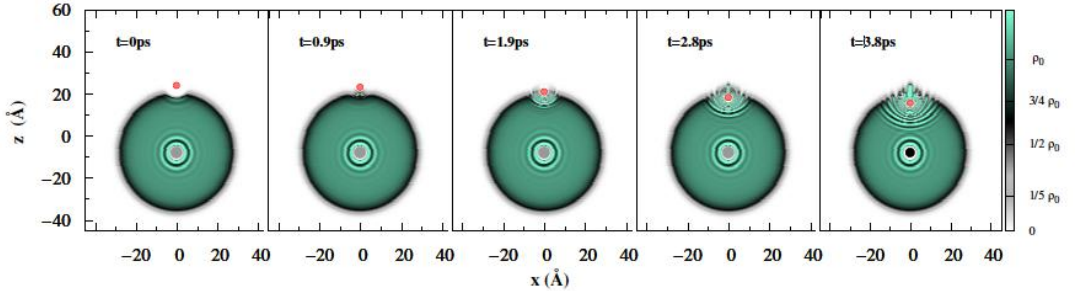

Figure 5: Snapshots every ≡ 0.95 ps taken during the solvation of Na^+ in a $^4He_{2000}$ droplet containing a Xe atom. At t = 3.8 ps (last snapshot) the Xe atom is ionized, triggering Coulomb explosion and the ejection of the ion with its solvation structure.

***References*:**

1. F. Ancilotto, M. Barranco, F. Coppens, J. Eloranta, N. Halberstadt, A. Hernando, D. Mateo, and M. Pi, *Int. Review in Phys. Chem.* (2017), **36**, 621.
2. S. H. Albrechtsen, C. A. Schouder, A. Viñas Muñoz, J. K. Christensen, C. E. Petersen, M. Pi, M. Barranco, and H. Stapelfeldt, Nature (2023), **623**, 319.
3. E. García-Alfonso, M. Barranco, N. Halberstadt, and M. Pi, J. Chem. Phys. (2024), **160**, 164308.

High-resolution configurational analysis of molecular adsorbates on rare-gas clusters

Arne Morlok, Philipp Elsässer, Ulrich Bangert, Yilin Li, Felix Riedel, Tanja Schilling, Frank Stienkemeier, and Lukas Bruder
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Rare-gas clusters are important model nanoreactors for multi-molecular chemistry [1,2]. The kinetics and reactivity of such systems are critically governed by the surface binding configurations of adsorbates and their thermal motion across the nanosurface. The associated dynamics can be complex, considering the thermal energy of the clusters being just below the boiling point and the large ensemble average of binding configurations, cluster sizes and morphologies. In particular, the morphology of the cluster surface and its potentially solid or liquid character may change with cluster size and upon transitioning from heavier to lighter rare-gas species. In order to disentangle this situation, we apply femtosecond two-dimensional spectroscopy which provides high temporal and spectral resolution beyond the ensemble average [3]. We use the method to reveal the ultrafast thermal hopping dynamics of phthalocyanine adsorbates on argon and neon cluster surfaces. Our results reveal the nanoconfinement of the adsorbates to specific surface facets. This has implications for multi-molecular reactivity in these systems but also confirms the solid character of the cluster surfaces.

References:

1. M. Briant et al., *Phys. Chem. Chem. Phys.* (2022), **24**, 9807
2. M. Fárník et al., *Phys. Chem. Chem. Phys.* (2021) **23**, 3195
3. U. Bangert, F. Stienkemeier, and L. Bruder, *Nat Commun* (2022) **13**, 3350.

Time-resolved photoelectron spectroscopy of organic molecules attached to rare-gas clusters

Frank Stienkemeier
Institute of Physics, University of Freiburg, Hermann Herder Str. 3, 79104 Freiburg, Germany

Superfluid helium nanodroplets and rare-gas clusters provide an ultracold environment down to millikelvin temperatures for the synthesis of molecular complexes and nanostructures. These systems enable detailed studies of charge and excitation transfer, along with the associated decay and loss mechanisms, allowing fundamental light-induced processes relevant to organic photovoltaics and optoelectronic devices to be isolated and understood. Our experiments combine fluorescence-based detection with photoionization and photoelectron spectroscopy. Real-time monitoring of laser-induced dynamics is achieved using femtosecond pump–probe techniques. In particular, time-resolved photoelectron spectroscopy offers direct insight into relaxation pathways following electronic excitation. Relaxation dynamics of excited acene molecules will be discussed, comparing results from molecular-beam experiments with those obtained under cluster-isolation conditions. Special emphasis will be placed on differences in spectroscopic signatures and in the temporal evolution of the underlying processes.

Simulating the Interstellar Medium in Helium Nanodroplets: From Molecular Formation to Spectroscopic Identification

Serge A. Krasnokutski[1], Anna Maria Reider[2], Lisa Ganner[2], Milan Ončák[2], Florian Foitzik[2], Stefan Bergmeister[2], Miriam Kappe[2], Fabio Zappa[2], Elisabeth Gruber[2], and Paul Scheier[2]

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Helium nanodroplets (HNDs) provide an exceptional platform for studying processes occurring in the interstellar medium (ISM). They replicate the extremely low temperatures of the ISM while offering isolated environments for species captured within the droplets. This presentation highlights two closely interconnected research directions: low-temperature chemistry and spectroscopy under such conditions.

Investigating chemistry at ultralow temperatures helps identify which species can form and persist in the ISM, thereby contributing to their observed abundances. Complementary spectroscopic studies yield experimental spectra that can be directly compared with astronomical observations, enabling the identification of these species in space. Such insights are crucial, as these molecules act as building blocks during planet formation and represent the initial stages of chemical evolution in the universe.

He-tagging spectroscopy has been employed to record UV–VIS–NIR spectra of several carbonaceous ions. The resulting absorption features were compared with diffuse interstellar bands (DIBs), which are a long-standing mystery in astronomy. As shown in Figure 1, the laboratory spectrum of C_4^+ exhibits a close match with the DIB at 503.9 nm [1]. Infrared spectroscopy further enables monitoring of reactions occurring inside HNDs [2] and provides a basis for comparison with unidentified infrared bands (UIBs).

Studies of low-temperature chemistry reveal efficient formation pathways for aminoketene and diketopiperazine (DKP) [3]. As illustrated in Figure 6, DKP^+ is predicted to exhibit exceptionally strong absorption in the visible range. Furthermore, DKP has also been detected in meteorites, consequently, DKP^+ is promising candidate for DIB carriers.

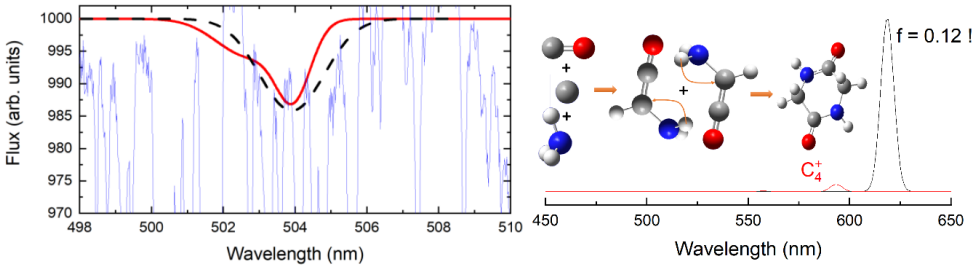

Figure 6: ***Left:*** *Comparison of laboratory and astronomical spectra. The solid red trace shows the attenuation of flux due to absorption by C_4^+. The solid blue trace represents the observational spectrum, while the dashed black trace corresponds to the DIB profile reported in previous studies.* ***Right:*** *Comparison of calculated spectra for C_4^+ (red) and DKP cations (black), together with the proposed mechanism for DKP formation in the ISM.*

References:

[1] S.A. Krasnokutski, L. Ganner, M. Ončák, F. Foitzik, S. Bergmeister, F. Zappa, P. Scheier, E. Gruber, , Astrophys. J., 982 (2025) 34.
[2] A.M. Reider, M. Kappe, M. Schmidt, F. Zappa, O.V. Lushchikova, S.A. Krasnokutski, P. Scheier, Mol. Phys. (2025) e2568076
[3] S.A. Krasnokutski, C. Jäger, T. Henning, C. Geffroy, Q.B. Remaury, P. Poinot, Sci. Adv., 10 (2024) eadj7179.

Fragmentation Dynamics and IR Spectroscopy of Acetonitrile–Formic Acid Clusters in Helium Nanodroplets

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Email: dmani@iitk.ac.in

Both acetonitrile and formic acid are of interest in interstellar chemistry [1–5]. Together, these molecules contain all four biogenically essential elements (C, H, N, and O). Therefore, their intermolecular complexes may represent intermediates in the formation of prebiotic compounds. These complexes were previously studied by Zims and Krim [6] in neon gas matrices. The 1:1 complex was found to have a cyclic structure stabilised by two hydrogen bonds.

In this work, we have generated (acetonitrile)$_m$–(formic acid)$_n$ complexes in helium nanodroplets using a recently developed helium nanodroplet isolation spectrometer in our laboratory [7,8]. The first goal of this study is to investigate the electron–impact–induced fragmentation dynamics of these complexes, and the second is to record mass-selective, high-resolution infrared spectra of these systems. Further details will be presented in the talk.

References:

1 https://cdms.astro.uni-koeln.de/classic/molecules.
2 S. B. Charnley, S. D. Rodgers, Y.-J. Kuan and H.-C. Huang, *Adv. Sp. Res.*, 2002, **30**, 1419–1431.
3 S. Cazaux, A. G. G. M. Tielens, C. Ceccarelli, A. Castets, V. Wakelam, E. Caux, B. Parise and D. Teyssier, *The Astrophysical Journal*, 2003, **593**:L51–L55, 2003.
4 A. Remijan, Y. Shiao, D. N. Friedel, D. S. Meier and L. E. Snyder, *Astrophys. J,* 2004, **617**, 384–398.
5 H. M. Boechat-Roberty, S. Pilling and A. C. F. Santos, *Astron. & Astrophys.*, 2005, **438**, 915–922.
6 E. L. Zins and L. Krim, *Phys. Chem. Chem. Phys.*, 2014, **16**, 3388–3398.

7 R. Gupta, S. Singha, D. Mani and Yashika, *Phys. Chem. Chem. Phys.*, 2025, **27**, 23865–23879.
8 D. Mani, R. Gupta, S. Singha, Yashika, P. Rani and P. Kumari, *J. Chem. Sci.*, 2026, **138**, 30.

Infrared Action Spectroscopy of Molecular Ions and Clusters in Helium Nanodroplets: Connecting Local Solvation Structure to Macroscopic Chemical Behavior

Daniel Thomas
University of Rhode Island, USA

To perform these studies, we will combine electrospray ionization mass spectrometry (ESI-MS) with helium-nanodroplet isolation infrared (HENDI IR) action spectroscopy to study size-selected, microsolvated ions and their complexes with gas-phase reactants. ESI-MS affords precise control over the investigated molecules, facilitating the precision isolation of target ion-water nanoclusters, which can be further modified via gas complexation within the mass spectrometer. Capture of carefully prepared, m/z-selected ions within a helium nanodroplet (equilibrium temperature of 0.4 K) enables the acquisition of high-resolution infrared spectra, revealing details of the molecular structure. In addition, the rapid cooling within the nanodroplet enables trapping of high-energy conformers populated or the stabilization of highly reactive species added to the droplet. These properties will be leveraged to enable mapping of complex potential energy surfaces associated with ion solvation and to capture pre-reactive complexes between microsolvated ions and reactive oxygen species.

This research will provide fundamental, molecular-level insights into the complex chemistry occurring at liquid/vapor interfaces, bridging the gap between theoretical models and macroscopic observations. Understanding interfacial reactions between ions and oxidants will enhance atmospheric models, improving predictions of atmospheric oxidizing capacity, aerosol composition, and radiative forcing, which impact air quality and climate. Furthermore, elucidating the mechanisms behind reaction acceleration and redox processes in microdroplets can inform the design of novel and efficient chemical synthesis strategies, including the conversion of inert species into valuable chemical feedstocks. Finally, a more comprehensive picture of the potential energy

surface of interfacial ion solvation will enable better predictions of ion reactivity in both microdroplets and aqueous aerosols.

Chemical reactions and structural dynamics in solution are governed by a complex interplay of both inter- and intramolecular interactions that can be challenging to disentangle. The transfer of molecules from solution to vacuum provides a complementary experimental approach that can pinpoint the properties of target systems. Our research group uses custom instrumentation that combines the gentle ionization and selectivity afforded by electrospray ionization mass spectrometry (ESI–MS) with the structural insight available from infrared action spectroscopy to interrogate species that are dynamic, transient, or otherwise difficult to isolate in solution. This presentation will provide an overview of the methodology and selected applications. In the analysis of biomolecules, we can provide a detailed description of the intramolecular interactions that give rise to the observed three-dimensional structure. However, the removal of solvent molecules can also dramatically alter the observed structure, and we are developing methods based on noncovalent complexation of charge recognition reagents to provide a "bridge" between solution and vacuum. Using ion spectroscopy, we are able to identify the binding motif and change in structure upon binding of a charge recognition reagent to the model peptide leucine enkephalin (YGGFL). We are also applying these techniques to identify intermolecular interactions crucial for liquid-liquid phase separation of short peptide sequences.

Electrospray ionization mass spectrometry (ESI-MS) enables the preparation of a wide range of analytes that can be extensively characterized under vacuum conditions using infrared (IR) ion spectroscopy. Various techniques for IR spectroscopy of ions have been developed, perhaps most commonly infrared photon dissociation (IRPD) action spectroscopy. In this approach, cluster dissociation is initiated by the sequential absorption of multiple resonant infrared photons. Ions may also be collisionally cooled to cryogenic temperatures and “tagged” with a messenger ion that is dissociated upon photon absorption. Alternately, helium nanodroplet isolation (HENDI) infrared ion action spectroscopy enables cooling of ions to 400 mK before spectroscopic characterization.

This work details the development of new instrumentation for both IRPD and HENDI IR action spectroscopy.

Electrospray ionization mass spectrometry (ESI-MS) allows for facile isolation and manipulation of molecular ions for further study under vacuum conditions, and coupling with infrared (IR) action spectroscopy facilitates investigation of molecular structure. Cooling of ions prior to spectroscopic characterization improves spectral quality by reducing the population of high-energy conformers or vibrational and rotational states. Capture of ions in superfluid helium nanodroplets enables cooling of entrained ions to 0.4 K with minimal matrix effects. Herein, we present a new helium nanodroplet apparatus for the capture of ions generated by ESI and stored in an ion trap.

Chemical reactions and structural dynamics in solution are governed by a complex interplay of both inter- and intramolecular interactions that can be challenging to disentangle. The transfer of molecules from solution to vacuum provides a complementary experimental approach that can pinpoint the properties of target systems. Our research group uses custom instrumentation that combines the gentle ionization and selectivity afforded by electrospray ionization mass spectrometry (ESI–MS) with the structural insight available from infrared action spectroscopy to interrogate species that are dynamic, transient, or otherwise difficult to isolate in solution. This presentation will provide an overview of the methodology and selected applications. In the analysis of biomolecules, we can provide a detailed description of the intramolecular interactions that give rise to the observed three-dimensional structure. However, the removal of solvent molecules can also dramatically alter the observed structure, and we are developing methods based on noncovalent complexation of charge recognition reagents to provide a "bridge" between solution and vacuum. We are also using this methodology to study the intermolecular interactions in deep eutectic solvents, which typically comprise a salt such as choline chloride and a hydrogen bond donor such as urea. These solvents are non-toxic, biocompatible, and have unique solvation properties. We are combining condensed-phase analysis approaches with infrared spectroscopy of isolated ionic clusters to connect global parameters to local interactions. This source provides a

continuous supply of droplets for coupling with a continuous-wave (cw) IR laser and quadrupole mass filter for ion action spectroscopy.

Macroscopic chemical properties and reactivity are fundamentally governed by local molecular interactions and solvation environments, yet directly connecting atomic-scale structural details to bulk-phase observations remains experimentally challenging. Here, we employ helium nanodroplet isolation infrared (HENDI) action spectroscopy coupled with electrospray ionization mass spectrometry (ESI-MS) to isolate and characterize size-resolved molecular clusters and ion-solvent complexes, providing a direct window into the solvation structures that dictate chemical behavior.

We will present results from several complementary systems demonstrating how this approach reveals the intermolecular interaction networks underlying complex solution chemistry. In deep eutectic solvents—multicomponent systems with tunable properties—we characterize isolated halide-molecule clusters to elucidate the competition between halide coordination and hydrogen-bonding networks. For biomolecular systems, we examine how noncovalent binding of charge recognition reagents provides a structural "bridge" between solution-phase and vacuum-phase conformations, revealing the energetic cost of solvation shell disruption. Additionally, we explore how infrared spectroscopy of isolated peptide clusters informs our understanding of intermolecular interactions governing liquid-liquid phase separation.

By comparing isolated cluster spectra with condensed-phase measurements and electronic structure calculations, we demonstrate how precise characterization of local solvation motifs connects to macroscopic observables. This integrated approach—combining the selectivity and control of ESI-MS with the structural detail afforded by cryogenic infrared spectroscopy—provides a versatile platform for understanding how solvation structure modulates chemical properties across diverse molecular systems.

Real-Time Probing of Hot Electron Diffusion in Superfluid Helium Nanodroplets by Laser-Assisted Electron Scattering

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The tracking of electron motion in real-time in nanostructures is a central challenge in nanotechnology and ultrafast science. It is demonstrated that hot electron propagation can be probed with femtosecond resolution using laser-assisted electron scattering (LAES), a light–matter interaction process in which electrons exchange energy with strong laser fields during collisions with atoms.[1,2]

Superfluid helium nanodroplets, doped with single atoms, are irradiated with pairs of few-cycle near-infrared laser pulses.[3] A quasi-free electron is released from the dopant via strong-field ionization by the first pulse, while its motion through the helium environment is probed by a time-delayed second pulse via LAES. The yield of accelerated electrons is found to directly reflect the propagation of hot electrons prior to localization into a bubble state. Comparison with Monte Carlo simulations indicates that electron motion follows a diffusive random walk with a diffusion coefficient of approximately 2 cm^2s^{-1}. Because elastic scattering constitutes the only energy-loss channel for 1–20 eV electrons in superfluid helium, the resulting high propagation length of several tens of nanometers establishes an upper limit for electron transport in materials under conditions where inelastic scattering is suppressed.

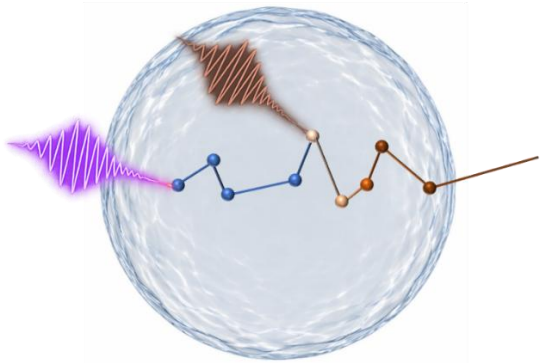

Figure 7: Schematic illustration of pump-probe laser-assisted electron scattering in a superfluid helium nanodroplet

***References*:**

1. L. Treiber, B. Thaler, P. Heim, M. Stadlhofer, R. Kanya, M. Kitzler, M. Koch, *Nat. Commun.* (2021), **12**, 4204.
2. L. Treiber, R. Kanya, M. Kitzler-Zeiler, M. Koch, *J. Phys. Chem. A* (2022), **126**, 8380-8387.
3. L. Treiber, Z. Wang, S. Ohrui, R. Kanya, M. Schultze, M. Koch, *in prep.* (2026)

From Superfluid Helium Nanodroplets to Functional Hybrid Nanomaterials

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Superfluid helium nanodroplets are widely used as ultracold, weakly perturbing hosts for cluster formation. This talk will focus on their use as a nanoscience platform: inert nanoscale reactors, soft-landing delivery media, and quantum-fluid environments in which weak interactions and quantized vortices can direct the growth of functional nanomaterials. Particular attention will be given to unusual deposited nanostructures that point to less-explored quantum-fluid behaviour during aggregation in superfluid helium.

The first part of the talk will discuss three materials-assembly routes enabled by helium nanodroplets. Sequential pickup of different vapours provides a direct route to core-shell nanoparticles and related heterostructures, with each component introduced independently during flight. Quantum vortices provide intrinsic one-dimensional templates, allowing trapped atoms and clusters to assemble into ultrathin nanowires without conventional surfactants, lithographic templates, or external fields. Molecular templates provide a further level of control: in the cold, inert helium environment, weak van der Waals interactions between metal atoms and specific binding sites on molecules become sufficiently influential to guide nanoparticle growth, enabling programmed nanoparticle assemblies with controlled interparticle spacing.

The second part will connect these assembly principles with functional materials. Examples will include plasmonic silver/gold nanostructures and hybrid metal-perovskite systems in which surface plasmon resonance

(SPR) is matched to the optical response of caesium lead bromide ($CsPbBr_3$). I will then discuss recent developments beyond conventional helium-droplet nanochemistry, including nanoparticle-programmed polymer surfaces for durable hybrid materials and protected strategies for producing high-quality perovskite structures. For these patent-associated developments, the presentation will focus on materials results and performance, not operational details. Overall, the talk will show how quantum-fluid cluster science can support controlled nanomaterial assembly, plasmon-enhanced optoelectronic structures, and technologically relevant hybrid interfaces.

Seeing double from a single shot: A two-color X-ray movie of superfluid helium nanodroplets

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The study of superfluid helium nanodroplets has been significantly advanced by Coherent Diffraction Imaging (CDI) at X-ray Free Electron Lasers (XFELs) [1]. CDI allows us to capture high-resolution images of isolated free-flying droplets, algorithmically retrieved from the X-ray diffraction signal [2]. This technique has revealed the intriguing behavior of dopants within these droplets [3], such as their tendency to agglomerate along quantized vortices, enabling their first direct visualization. Thanks to the extensive knowledge built over the last decades and the consequent level of control over droplet size and dopant structure [4], helium nanodroplets now represent the ideal reference system for the study of light-matter interaction and proof-of-concept experiments. In this regard, XFELs are capable of producing pairs of ultrashort light pulses of different wavelengths with controllable sub-picosecond time delays, which have recently reached sufficient brightness to yield a diffraction pattern. However, the two diffraction signals from the two pulses overlap on the detector, preventing conventional CDI analysis from retrieving the individual images [5]. We have recently introduced a new framework, Dichography, to handle this new class of diffraction experiments [6]. Dichography renders the overlapping of the two scattering signals tractable and allows for the reconstruction of the two corresponding images of the sample. This methodological advancement is supported by the first experimental demonstration of the technique, where two time-delayed images of a single helium nanodroplet and its inner dopants are reconstructed. Such images are, in every sense, a two-frame movie of the sample, and open new, exciting possibilities for the time-resolved visualization of ultrafast processes at the nanoscale.

***References*:**

1. O. Gessner and A. F. Vilesov, Imaging quantum vortices in superfluid helium droplets, *Annual review of physical chemistry* (2019) **70**, 173.
2. A. Colombo and D. Rupp, Imaging Clusters and Their Dynamics with Single-shot Coherent Diffraction. *Structural Dynamics with X-ray and Electron Scattering* **25** (2023) 172-232.
3. R. M. P. Tanyag, C. F. Jones, C. Bernando, S. M. O. O'Connell, D. Verma, and A. F. Vilesov, Experiments with Large Superfluid Helium Nanodroplets, in *Theoretical and Computational Chemistry Series, Royal Society of Chemistry* (2017) 389-443.
4. A. Ulmer, et al., Generation of large vortex-free superfluid helium nanodroplets, *Physical review letters* (2023) **131**, 076002.
5. L. Hecht, et al., Two-Color X-Ray Coherent Diffraction Imaging of Helium Nanodroplets. *arXiv preprint arXiv:2508.19991* (2025).
6. L. Hecht, et al., Dichography: Two-frame Ultrafast Imaging from a Single Diffraction Pattern. *arXiv preprint arXiv:2508.20153* (2025).

Solving atomic structures of nanoclusters inside superfluid helium droplets

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Nanoclusters inside superfluid helium droplets can exhibit esoteric structures due to the quantum solvent. Deposition of these clusters onto transmission electron microscope (TEM) grids can induce structural changes. Electron diffraction of these clusters prior to deposition offers an in situ method for structure determination. However, the surrounding helium atoms pose a strong background, masking the interferences due to the atoms of the cluster. We recently succeeded in introducing rare atoms as a contrasting agent for the embedded nanoclusters, and observed strong interferences from the cluster complex. This approach is similar to the negative staining experiments in TEM. I will present results from these experiments, using a few organic molecules and xenon atoms as examples. We also report complications due to “foaming” of argon atoms as the contrasting agent.

Ultrafast and ultraslow relaxation of excited He nanodroplets

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Helium nanodroplets are often considered the ultimate matrix for spectroscopy of isolated molecules, molecular complexes and even nanostructures that are hard to form by any other means. However, when He nanodroplets are photo-excited or ionized, they themselves turn into highly reactive species that feature intriguingly complex relaxation and reaction dynamics.

Using extreme ultraviolet (XUV) light sources such free-electron lasers (FEL) and high-harmonic generation (HHG) sources based on intense femtosecond lasers that have recently become readily available, we probe the ultrafast relaxation dynamics of pure and doped He nanodroplets triggered by ultrashort XUV pulses. In this way, we obtain information about how a superfluid reacts to impulsive perturbation in time and how fast it interacts with a dopant in or on the droplet. It turns out that He nanodroplets are excellent model systems to study fundamental radiochemical processes that are also important in radiation biology, such as secondary electron production, elastic and inelastic electron scattering, and intermolecular transfer of energy and charge. Special features of He nanodroplets are the formation of metastable excited He* atoms which migrate to the droplet surface and roam along the surface, and the transfer of energy from one He* atom to another or from a He* to a dopant attached to the same droplet in a so-called interatomic Coulombic decay (ICD) process.

Over the past years, we discovered various types of ICD processes prevailing in He nanodroplets excited or ionized in different regions of the XUV spectrum [1]. In my talk, I will present recent results on the relaxation dynamics of XUV-excited and ionized He nanodroplets, including ultrafast

[2] and ultraslow [3] ICD of pairs of He* excitations probed by laser-induced XUV fluorescence [3] or electron spectroscopy, see Fig. 8.

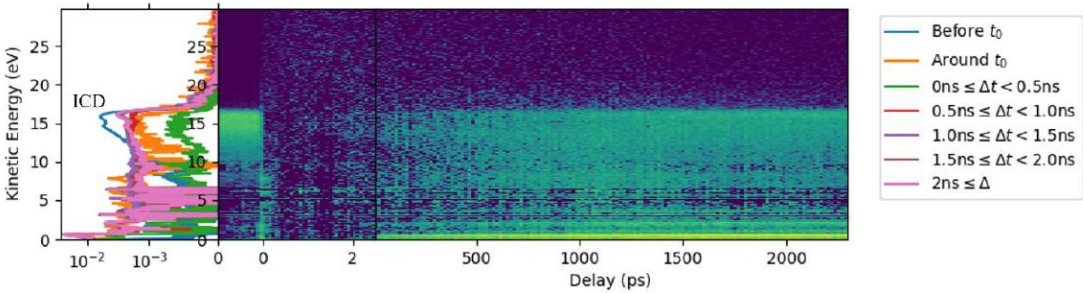

Figure 8: Time-resolved electron spectra of He nanodroplets resonantly excited by an XUV pump pulse and probed by an intense NIR pulse. The peak at 15 eV, which is depleted at short pump-probe delays 0-2 ps, is due to interatomic Coulombic decay (ICD) of pairs of excited He atoms, $He^* + He^* \rightarrow He + He^+ + e_{ICD}$.

References:

[1] A. C. LaForge et al., Rep. Prog. Phys. (2024), 87, 126402
[2] A. C. LaForge et al., Phys. Rev. X (2021), 11, 021011
[3] K. Sishodia et al., Sci. Rep. (2026), **16, 1321**

Transitional supersolidity in Helium droplets

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If a helium droplet is doped with an alkali ion, it turns into a snowball with some crystallized helium shells in the interior, surrounded by superfluid helium atoms. In between forms a layer which is supersolid, i.e, has localized density of helium atoms, which, however, behave coherently like a superfluid [1].
This is a surprising example of supersolidity in helium, a strongly interacting system. In fact supersolidity was originally proposed for bulk helium, but to date it has not been clearly identified experimentally.
In the second part of the talk I will add a Rydberg electron and determine its properties circling the liquid ionic core. Its peculiar spectral features may be used to identify the snowball structure experimentally [2]. We can treat relatively large droplets which are out of reach for quantum Monte Carlo techniques thanks to a self-consistent hybrid approach of Gaussians, describing the crystallized atoms, and a suitable DFT treatment for the superfluid part [3].

References:

[1] JC Matos, P Giannakeas, M Cardi, T Pohl, JM Rost, Phys. Rev. Lett. 134, 186001 (2025)
[2] JC Matos, P Giannakeas, JM Rost, submitted (2026)
[3] JC Matos, PhD thesis, Dresden (2025)

Quantum Microsolvation of H_2^+ and D_2^+ in Helium Clusters: Data-Driven Potentials, Growth Patterns and Nuclear Quantum Effects

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Helium nanoclusters doped with protonated impurities offer a unique environment to study quantum microsolvation, where weak He-He binding, strong nuclear delocalization, and anisotropic ion-He forces combine to produce highly structured solvation shells. Here we report a computational study of the microsolvation of H_2^+ in helium clusters, $H_2^+He_N$, for N up to 30 [1,2]. Data-driven He-H_2^+ interaction potentials trained on CCSD(T)/CBS ab initio data are incorporated into a sum-of-potentials framework and used in basin-hopping optimization as well as classical and semiclassical Feynman-Hibbs (FH2) molecular dynamics at T = 2 K [1]. The calculations reveal distinct solvation microstructures centered on a persistent $[He\text{-}H\text{-}H\text{-}He]^+$ core, with helium rings growing outward, and single-atom evaporation energies are compared with experimental ion-yield data [3]. Complementary Rigid Body Diffusion Monte Carlo (RBDMC) simulations [2] provide exact ground-state energies and 3D probability densities for both $H_2^+He_N$ and $D_2^+He_N$, showing that the inner two helium atoms remain strongly localized while outer shells display coexisting solid-like and liquid-like character. Quantum delocalization and He-exchange in the outer shells prevent the emergence of clear magic-number structures, and isotopic substitution (H_2^+ vs. D_2^+) produces measurable structural differences for $N \geq 10$. The results point to the need for higher-order nonadditive terms in the potential model and

for contamination-free high-resolution mass spectra to guide future refinement.

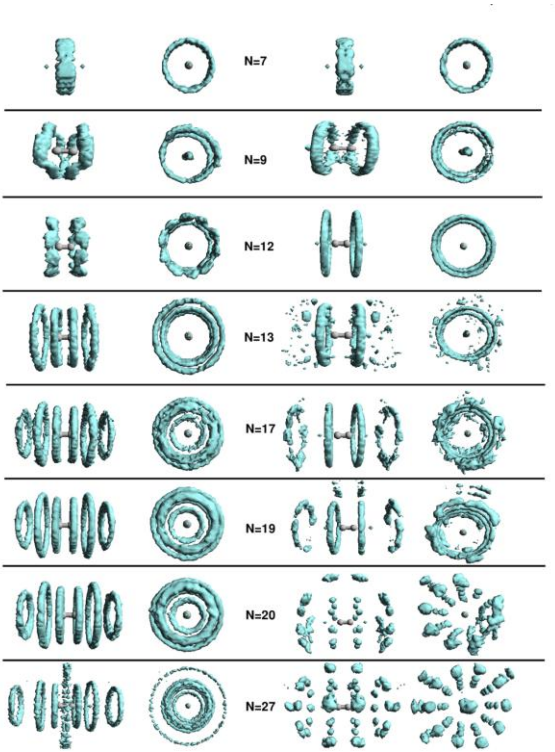

Figure 9: 3D spatial probability distributions of He atoms around the H_2^+ cation from classical MD (left columns) and semiclassical FH2 (right columns) simulations for selected $H_2^+He_N$ clusters at T = 2 K [1].

***References*:**

1. M. J. Montes de Oca-Estévez, P. Villarreal, J. Hernández-Rojas, J. Bretón, A. Sarsa and R. Prosmiti, *Chem. Phys. Lett.*. (2025), **877**, 142242.
2. A. Sarsa, M. J. Montes de Oca-Estévez, P. Villarreal, J. Hernández-Rojas and R. Prosmiti, *Phys. Chem. Chem. Phys.* (2026), **28**, 5369–5378.
3. P. Bartl, C. Leidlmair, S, Denifl, P. Scheier and O. Echt, *Chem. Phys. Chem*. (2013), **23**, 181-185.

Hydrogen attachment to Na-decorated polycyclic aromatic hydrocarbons: a theoretical approach in comparison with helium nanodroplets experiments

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Functionalizing carbon-based materials with metal atoms has been proposed as an effective means to enhance the adsorption of hydrogen molecules on these substrates, thereby developing new useful systems for hydrogen storage. With this goal in mind, our group has been carrying out some theoretical work on the attachment of H_2 molecules to substrates of increasing complexity, in close connection with helium nanodroplets (HND) experiments carried out by the Scheier's group in Innsbruck[1-3]. In particular, we build reliable H_2-substrate and H_2-H_2 interaction potentials based on accurate *ab initio* calculations and, using these force fields, we obtain the ground states of clusters with several H_2 molecules adsorbed on the substrates by means of Quantum Monte Carlo methods. The approach has been applied to the attachment of hydrogen to Na^+[1] as well as to (charged) bare and Na-decorated polycyclic aromatic hydrocarbons (naphthalene[2] and coronene[3]). It is found that some cluster sizes (number of molecules attached) exhibit relatively large evaporation energies, therefore being more stable. These findings are in general in quite good agreement with the observed abundances of the ions produced inside the HNDs. In addition, it has been clearly found from the experiments -and supported by the calculations- that a Na-decorated support retains a considerably larger number of molecules than the undecorated one, in agreement with an enhancement of the hydrogen adsorption due to the role of the decorating metal. The results prove that HNDs are a useful tool for these adsorption studies.

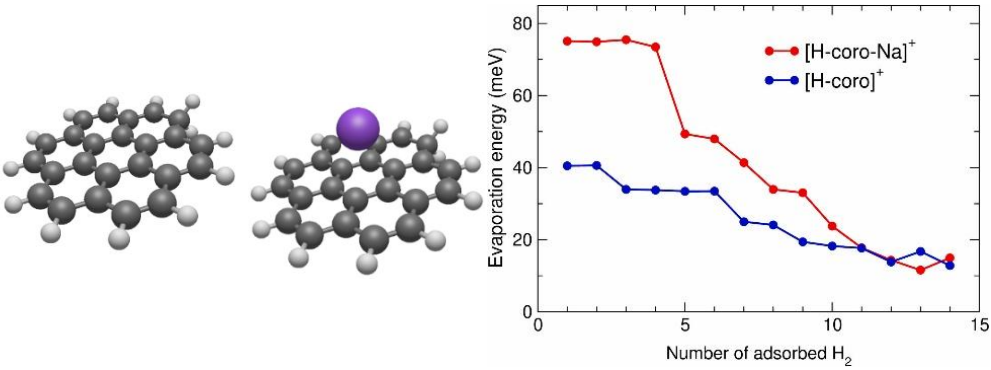

Figure 10: Bare and Na-decorated protonated coronene and enhancement of H_2 evaporation energies in the decorated substrate.

***References*:**

1. S. Kollotzek, J. Campos-Martínez, M. Bartolomei, F. Pirani, L. Tiefenthaler, M. I. Hernández, T. Lázaro, E. Zunzunegui-Bru, T. González-Lezana, J. Bretón, J. Hernández-Rojas, O. Echt, P. Scheier. *Phys. Chem. Chem. Phys*. (2023), **25**, 462-470.

2. A. M. Reider, S. Kollotzek, P. Scheier, F. Calvo, E. Yurtsever, F. Pirani, M. Bartolomei, M. I. Hernández, T. González-Lezana, J. Campos-Martínez**.** *Int. J. of Hydrog. Energy* (2024) **58**, 525-535.

3. E. García-Arroyo, A. M. Reider, S. Kollotzek, F. Foitzik, J. Campos-Martínez, M. Bartolomei, F. Pirani, M. I. Hernández, M. Mella, P. Scheier, *Int. J. Hydrog. Energy* (2024), **83**, 387-395.

Quantum Rotation in Helium Nanodroplets

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This talk reviews our studies of isolated sub-micrometer droplets of superfluid ^{4}He, normal fluid ^{3}He, and mixed systems, investigated using single-shot femtosecond X-ray coherent diffractive imaging at free-electron laser facilities. The droplets carry substantial angular momentum, leading to pronounced centrifugal deformation. To visualize vortices in superfluid ^{4}He droplets, Xe atoms are introduced; these preferentially accumulate in vortex cores, forming thin filamentary structures. In contrast, in ^{3}He droplets, Xe atoms assemble into diffuse, ring-shaped distributions along the equatorial region.

The equilibrium shapes of rotating superfluid droplets span a sequence of oblate and prolate geometries, evolving along stability curves parameterized by reduced angular momentum and angular velocity. In axisymmetric oblate droplets, angular momentum is entirely accommodated by triangular lattices of quantized vortices. In prolate configurations, however, capillary waves contribute significantly—and for sufficiently large aspect ratios, can even dominate—the angular momentum. Additional topics include quantum phase separation in mixed ^{3}He/^{4}He droplets and the arrangement of multiple charges on droplet surfaces.

Hot Topic Talks

Observation of the metastable He tetramer cations He_4^+ in helium droplets

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Electron-impact ionization of helium nanodroplets initiates various processes owing to their intrinsic properties: generation of fragment helium cluster cations and dopant cations, realization of multiply charged droplets [1], and stabilization of molecular cations in the droplets [2]. In the fragment mass spectrum, it is known that the anomalously intense He tetramer cation He_4^+ peak appears when the droplet size is in the range of $\langle N_{He} \rangle \geq 105$. This is attributed to the formation of metastable He_4^{+*} in the quartet spin state, where its production mechanism has been discussed thoroughly [3]. The distinct visible absorption band of the He_4^{+*} fragment cation was observed experimentally [4] and supported theoretically by ab-initio calculations [5]. In this study, we explore the possibility of its existence in the droplet as a charge carrier.

We produced helium nanodroplets using a cryogenic pulsed nozzle operating at a repetition rate of 10 Hz. The nozzle condition was set to a pressure of 2 MPa in a temperature range of 7 to 20 K, corresponding to the droplet size of $\langle N_{He} \rangle$ = 1010 to 103. After electron-impact ionization, the positively charged droplets traversed to the acceleration region of a time-of-flight mass spectrometer (TOF-MS) on the millisecond timescale; in this way, we separated the charged droplets from the fragment cluster cations. A visible nanosecond pulsed laser was used to excite the charged droplets in the TOF-MS acceleration region, enabling mass-selective detection of photo-released cluster cations.

Figure 11 shows an observed TOF-MS spectrum at an excitation wavelength of 620 nm, where He_2^+ and He_4^+ are predominant and He_3^+ is notably absent, both of which are expected from the He_4^{+*} structure (Fig. 1 inset). The He_4^{+*} appearance threshold in electron energy at 40 eV and the linear laser energy dependence of the He_4^{+*} intensity strongly support the existence of this metastable cation in the droplets. The narrow spectrum in the droplet, compared to that observed previously [4], suggests that the vibrational energy of this ion is cooled within 1 ms.

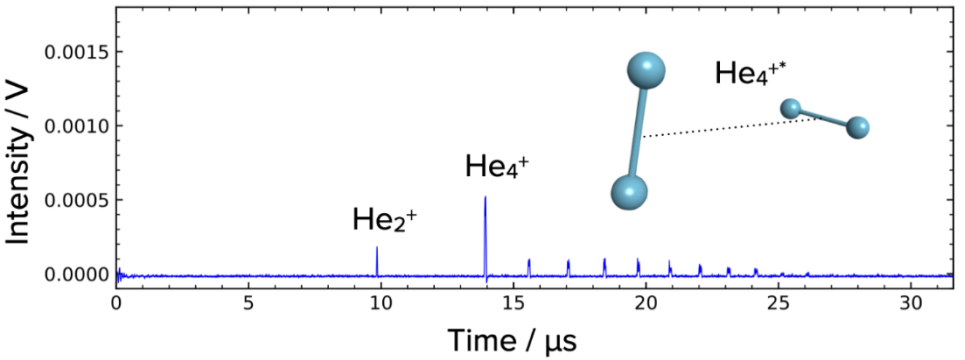

Figure 11: TOF-MS spectrum of helium droplets ($\langle N_{He} \rangle = 10^7$) irradiated with a nanosecond pulsed laser at 620nm measured 1 ms after electron impact ionization at 100 eV. Inset: the calculated He_4^{+*} structure

***References*:**
[1] F. Laimer et al., *Phys. Rev. Lett.* **123**, 165301 (2019).
[2] D. Verma et al., *J. Phys. Chem. A* **124**, 6207 (2020).
[3] J. Fine et al., *J. Chem. Phys.* **148**, 044302 (2018). [4] B. von Issendorff et al., *Chem. Phys. Lett.* **233**, 23 (1995).
[5] P. J. Knowles and N. Murrell, *J. Chem. Phys.* **102**, 9442 (1995).

Liquid-like Quantum Matter in Multicomponent Bose Gases

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Recent theoretical results will be presented on ultracold binary mixtures of bosonic atoms with tunable interactions, which provide a versatile platform to explore phenomena such as quantum droplets, mixed phases, and topological superfluid structures (rings, shells).
These results are based on numerical simulations using extended Gross-Pitaevskii equations including quantum fluctuations, applied to ^{41}K-^{87}Rb mixtures in the regime where a sufficiently attractive interspecies interaction leads to the formation of self-bound, liquid-like quantum droplets. These quantum droplets, first predicted theoretically[1] and subsequently observed experimentally in dipolar gases[2], homonuclear[3] and heteronuclear[4] binary mixtures of bosonic atoms, are superfluid systems characterized by ultralow equilibrium densities and a finite (albeit extremely small) surface tension, and therefore exhibit liquid-like properties while maintaining quantum coherence. These make them a unique form of matter that bridges the gap between classical liquids and quantum gases[5].
A more recent development will also be discussed, in which a three-component Bose mixture made of ^{23}Na, ^{39}K and ^{41}K atoms at ultralow temperatures (species 1,2,3 in the following) is studied [6,7].
The binary mixtures (1,2) and (2,3) can each separately form quantum droplets. A possible, low-energy configuration for the three species mixture is a structure where two quantum droplets (made of the (1,2) fluid and (2,3) fluid, respectively) are bound together by the shared component 2, forming a stable “quantum dimer” structure, which displays vibrational modes surprisingly similar to those of a classical diatomic molecule. A simple protocol is proposed to create a stable linear chain formed by

periodic repetition of this basic building block, i.e., an alternating sequence of (^{23}Na,^{39}K) and (^{39}K,^{41}K) droplets. This structure exhibits both periodic density modulations
from the droplet ordering, and global phase coherence due to the shared ^{39}K component, satisfying the criteria for supersolidity. I will illustrate some properties of this supersolid "diatomic linear chain", which represents an interesting system bridging cold-atom physics with condensed-matter properties[8].

***References*:**

1. D. S. Petrov, *Phys. Rev. Lett. (2015),* ***115****, 155302.*
2. M. Schmitt et al., *Nature (2016),* ***539****, 259;* L. Chomaz et al., *Phys. Rev. X (2016),* ***6****, 041039.*
3. C.R. Cabrera et al., *Science (2018),* ***359****, 301.*
4. C. D'Errico et al., *Phys.Rev. Res. (2019),* ***1****,* 033155.
5. M. Caldara and F. Ancilotto, *Phys. Rev. A (2022),* **105**, 063328
6. Y. Ma and X. Cui, *Phys. Rev. Lett. (2025),* ***134****,* 043402.
7. F. Ancilotto, *Phys. Rev. Lett. (2025),* ***135****,* 159301.
8. F. Ancilotto, *Phys. Rev A* (2025), **112**, 063317.

Electrostatic Trapping of Multiply Charged Helium Nanodroplets

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A Multi-Reflection Ion Beam Trap device, developed in collaboration between the TU Darmstadt and the University of Greifswald[1], is used to confine multiply charged helium nanodroplets (HNDs) for up to minute-long timescales using purely electrostatic fields. Proof-of-principle measurements demonstrate efficient storage, with droplets completing thousands of revolutions. The storage time strongly depends on the background pressure and has been systematically investigated.
Investigation of Blackbody Infrared Radiative Decay (BIRD) of water doped HNDs, utilizing the chamber as a blackbody emitter, reveal, that vibrations of embedded water clusters lead to enhanced helium evaporation, eventually reducing the droplet's kinetic energy below detection limits.[2]
Additionally, the planned implementation of a pick-up electrode will allow investigation of temporal changes in charge, mass, and energy, providing a new approach to nano-calorimetry and enabling thermodynamic studies at the nanoscale. The development of image charge detection is motivated by pioneering measurements of Hanozin et al.[3], which observed significant charge loss and Rayleigh decay in aqueous droplets.

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***References*:**
[1] Schlaich, M.; Fischer, J.; Fischer, P.; Klink, C.; Obertelli, A.; Schmidt, A.; Schweikhard, L.; Wienholtz, F. A Multi-Reflection Time-of-Flight Mass Spectrometer for the Offline Ion Source of the PUMA Experiment. *Int. J. Mass Spectrom.* **2024**, *495*, 117166.
[2] Veternik, M.; Waldhütter T.; Schweikhard L.; Scheier P.; Gruber E. Extending

the Observation Time of Charged Helium Droplets to the Minute Timescale. *Phys. Rev. Lett.* **2026**, 136, 013201.
[3] Hanozin, E.; Harper, C. C.; McPartlan, M. S.; Williams, E. R. Dynamics of Rayleigh Fission Processes in ∼100 Nm Charged Aqueous Nanodrops. *ACS Cent. Sci.* **2023**, *9* (8), 1611–1622.

Cryogenic Bottom-up Formation of the Benzene Cation from Acetylene in Helium Nanodroplets

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Ion-molecule reactions, often characterized by very low or vanishing activation barriers, play a fundamental role in the formation of complex organic molecules in space. One key molecular precursor of larger aromatic molecules is acetylene due to its unique stability and reactivity upon ionization. This molecule is one of the most promising candidates for bottom-up processes, which lead to the formation of polycyclic aromatic hydrocarbons [1]. Nevertheless, despite extensive efforts [2], a viable formation pathway for even the simplest aromatic building block, benzene, under astrophysical conditions has not yet been confirmed. In fact, one promising pathway, starting with the protonation of acetylene, was cast into doubt recently [3].

In this contribution, we report the formation of the benzene radical cation, formed by the sequential addition of two neutral acetylene molecules to the acetylene cation. The reaction was observed inside helium nanodroplets at cryogenic temperatures. The product is unambiguously characterized as benzene through helium tagging spectroscopy in the UV/vis spectral region and by the investigation of its solvation characteristic in neutral acetylene [4].

***References*:**

1. Pentsak et al., *ACS Earth Space Chem.* **8**, 798 (2024)
2. Geppert et al., *Chem. Rev.* ***113***, 8872 (2013)
3. Kocheril et al., *Nat. Astron.* **9**, 685 (2025)
4. Foitzik et al., *J. Am. Chem. Soc.*, **148**, 18615 (2026)

Insight into electron-beam-induced nano-manufacturing from cluster-beam experiments

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In recent years, a lot of effort has been dedicated into understanding electron-induced chemistry of organometallic molecules. The motivation comes from use of such compounds as precursors in focused-beam nano-manufacturing. There, metal-ligand bonds are cleaved by a focused electron beam and this enables 3D nanoprinting of metallic structures. Numerous attempts have been made to correlate the deposit metallic content with fragmentation patterns of gas-phase precursor molecules upon their interaction with the electrons. The gas-phase experiments of course cannot capture the effect of the environment – especially of the substrate – on such reactions.

I will demonstrate how cluster-beam experiments can be used to mimic the presence of the substrate. We use mainly large argon clusters on which we pick-up precursor molecules. Already such an elementary environment has profound effects on the outcome of electron-induced chemistry. The fragmentation degree in dissociative ionization is strongly suppressed upon clustering [1]. As to the dissociative electron attachment, the low-energy loss of one ligand, which dominates the gas-phase spectrum, is completely absent in clusters [2]. Recently these effects have been put on quantitative basis using irradiation-driven molecular dynamics simulations [3]. Such simulations additionally enable us to quantify the expected differences between precursors on clusters and on the substrates relevant in nano-manufacturing conditions [4].

Since the experiments described above do not involve any quantum-fluid clusters, a small part of my contribution will be dedicated to the

description of a new helium-nanodroplet setup which is currently being built in our laboratory.

***References*:**

1. J. Lengyel et al., *J. Phys. Chem. C* (2016), **120**, 17810, doi: 10.1021/acs.jpcc.6b05852
2. J. Lengyel et al., *Beilstein J. Nanotechnology* (2017), **8**, 2200, doi:10.3762/bjnano.8.219
3. B. Andreides et al., *J. Phys. Chem. A* (2023), **127**, 3757, doi: 10.1021/acs.jpca.2c08756
4. H. Lyshchuk et al., *J. Phys. Chem. C* (2026), under review

Observing time-resolved stereodynamics in bimolecular cation-molecule reactions inside liquid helium droplets

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Recently, experimental results have shown that it is possible to measure diffusion-limited bimolecular reactions in real-time using liquid helium nanodroplets doped with a lithium atom and a benzene molecule, pump-probe femtosecond spectroscopy and ion-coincidence velocitymap imaging [1]. Here, that method is extended to substituted aromatic systems, which allows for 1) varying the electrostatic properties of the molecule and 2) using Coulomb Explosion Imaging to probe the orientation of the molecule relative to the cation.

For 1,3,5-trichlorobenzene, a quadrupole-like system, Li^+ forms a complex with the molecule during the first 100 ps, where Li^+ binds out-of-plane. This is determined by considering the relative recoil directions between Li^+ and Cl^+ ions as shown in Fig. 12.

For 4-fluoroiodobenzene, a dipole-like system, Li+ binds along the F-C bond, showing a 180° relation between recoiling Li+ and I+ ions and no relation between Li^+ and F^+ ions.

Finally, for 4-chloroiodobenzene, Li^+, I^+ and Cl^+ ions are all observed recoiling together, indicating that Li^+ binds out-of-plane but tilted towards the I-substituent. By simultaneously measuring Li^+He_n ions, a shift in the recoil direction of Li^+He_n ions with I+ as the cation enters the droplet can be observed, changing from the long-range dominated dipole configuration (with Li^+He_n along the Cl-C bond) to the complex close-range interaction (with Li^+ binding out-ofplane). The dynamics show that this reorientation is limited by the droplet size distribution, taking place over 30-100 ps.

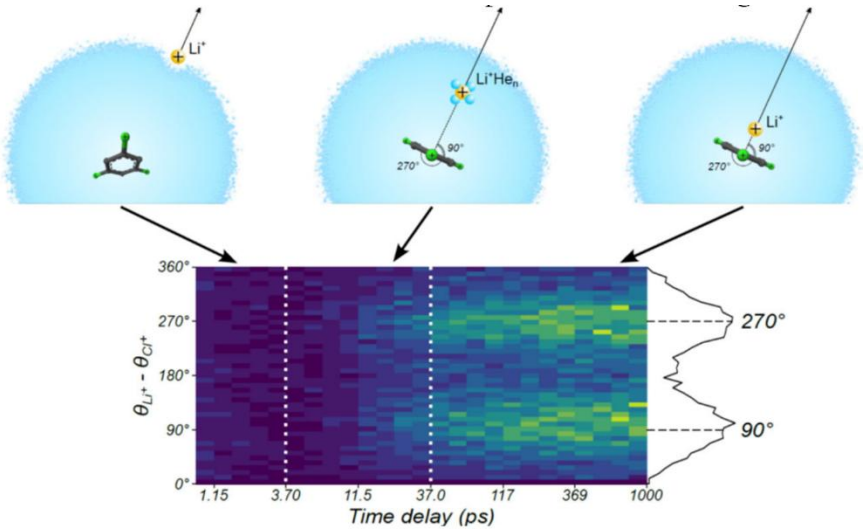

Figure 12: Schematic of reaction and time-resolved covariance map trace of the recoil angle between Li_+ and Cl_+ ions

***References*:**

1. J. K. Christensen, C.E. Petersen, S.H. Albrechtsen *et al.*, *Nat Commun* (2026), **17**, 1249

Investigating rotational decoherence with non-adiabatic laser-induced alignment of Li_2 molecules on superfluid helium nanodroplets

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It has recently been demonstrated that alkali metal dimers residing on the surface of superfluid helium nanodroplets can be driven into coherent rotation. These studies revealed that (i) the rotational dynamics are largely confined to the tangent plane of the droplet surface, effectively yielding two-dimensional motion with a significantly reduced rotational constant, and (ii) the degree of alignment decays on a timescale of a few hundred picoseconds[1]. Such behaviour has been observed for Na_2, K_2, and Rb_2 in both singlet and triplet states[2].

In this work, we present a detailed investigation of the laser-induced, non-adiabatic rotational dynamics of Li_2 molecules on helium nanodroplets. Specifically, we examine the time-dependent degree of alignment across a range of kick-pulse intensities. Increasing the pulse intensity leads to a higher prompt alignment immediately following the kick, accompanied by a narrowing of the alignment peak, indicating the population of higher rotational states.

More intriguingly, within the first few tens of picoseconds after the prompt alignment, we observe that - independent of pulse intensity - the rotational dynamics are dominated by a single coherence: the lowest-energy coherence between the $J = 0$ and $J = 2$ states. The mechanism underlying this behavior remains unclear. We explore possible explanations, including coupling to surface excitations of the droplet (ripplons) and interactions with bulk modes[3].

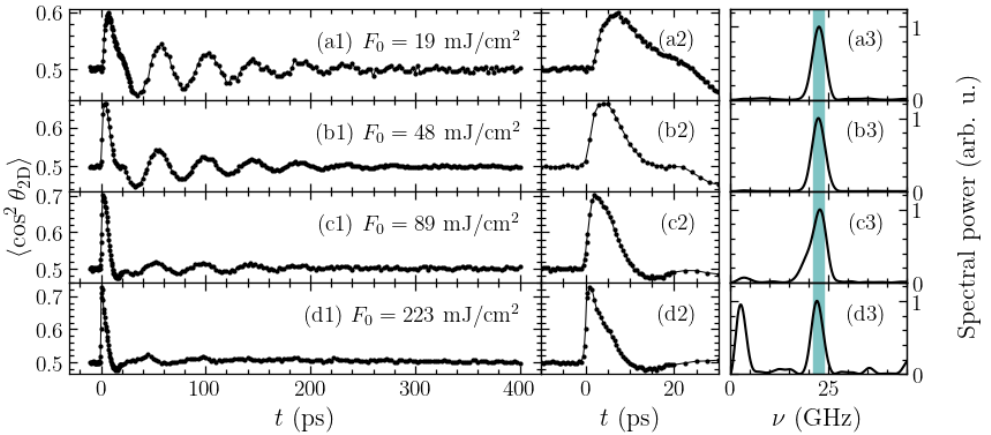

Figure 13: (a1)-(d1): Measured degree of alignment for a selection of kick pulse fluences. (a2)-(d2): Zoom-ins on the early dynamics. (a3)-(d3): Fourier transforms of the alignment traces revealing a single coherence.

References:

1. L. Kranabetter, H. H. Kristensen, et. Al., *Phys. Rev. Lett.* (2023), **131**, 053210.
2. H. H. Kristensen, L. Kranabetter, et. Al., *Phys. Rev. A.* (2025), **111**, 033114.
3. K. Hansen, M. D. Johnson, V. V. Kresin, *Phys. Rev. B.* (2007), **76**, 235424.

PIConGPU modeling of nanoplasma formation in helium nanodroplets irradiated by intense femtosecond laser pulses

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Helium nanodroplets provide a unique and versatile platform for investigating strong-field-driven nanoplasma dynamics[1]. In this work, we present large-scale, GPU-accelerated particle-in-cell simulations using PIConGPU[2] to study the interaction of pure helium nanodroplets containing up to 10^6 atoms irradiated with a 1e15W/cm^2 intense near-infrared femtosecond laser pulses, and we compare the results with single-shot velocity-map electron imaging and ion measurements. The simulations describe the plasma evolution from the first ionization events to collective electron motion, nanoplasma formation, and early expansion.

We show that the calculated electron and ion observables reproduce the main features of the measured spectra in systems with similar cluster sizes and laser intensities. Our results demonstrate that PIConGPU captures the essential physics of nanoplasma formation previously addressed mainly with molecular-dynamics or TDDFT approaches [3], while remaining computationally efficient and applicable to much larger systems. This establishes PIConGPU as a powerful and scalable tool for connecting nanoplasma theory with experimentally accessible observables.

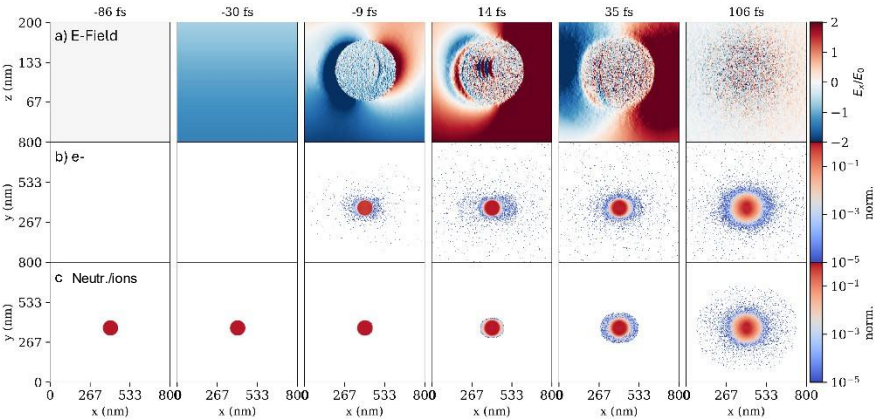

Figure 14: Spatio-temporal evolution of a laser-driven helium nanoplasma for the initially neutral target. a) normalized laser field component in the polarization direction plane (polarization along x, propagation along z). b): electron distribution and c) helium core-particle distribution (all charge states, Z = 0, 1, 2). Times are given relative to the laser peak at the target (t = 0)

References:

1. A. Mikaberidze, "Laser-Driven Nanoplasmas in Doped Helium Droplets: Local Ignition and Anisotropic Growth," Physical Review Letters, 2009.

2. "PIConGPU: Particle-in-Cell Simulations for the Exascale Era." https://github.com/ComputationalRadiationPhysics/picongpu, 2026. [Online; latest release picongpu-0.8.0 at 18-December-2024].

3. C. Peltz, C. Varin, T. Brabec, and T. Fennel, "Fully microscopic analysis of laser-driven finite plasmas using the example of clusters," New Journal of Physics, 2012.

Time-resolving interatomic Coulombic decay in He nanodroplets using nanoplasma flourescence

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Superfluid helium nanodroplets that are resonantly excited using extreme ultraviolet (XUV) pulses can relax through interatomic Coulombic decay (ICD)1-3 , a process typically regarded as ultrafast 3, 4. In this work, we present a new method to investigate ICD dynamics5 in helium nanodroplets across a wide time range, from femtoseconds to nanoseconds. The approach is based on measuring XUV fluorescence emitted when the droplets are transformed into a nanoplasma by subsequent intense infrared pulses. The formation of this nanoplasma is triggered by tunnel ionization of XUV-excited helium atoms bound to the droplets6, making it a sensitive indicator of their relaxation behavior. The observed decay on the nanosecond timescale is attributed to ICD occurring between pairs of excited helium atoms that move across the droplet surface, a mechanism supported by both quantum-mechanical and classical modeling.

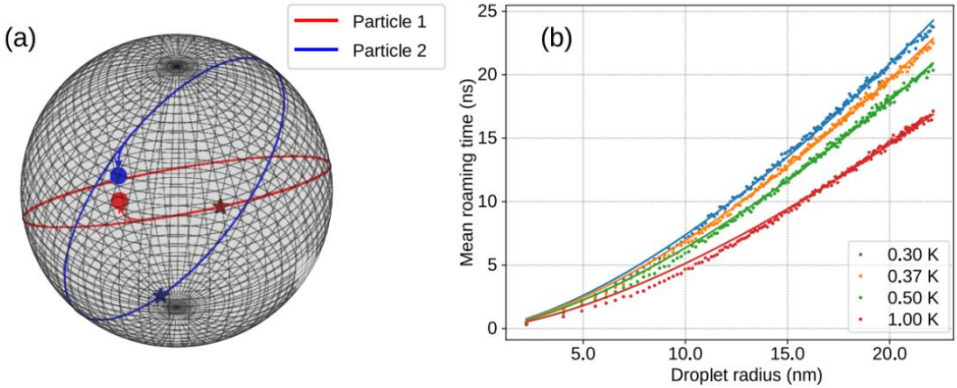

Figure 15:(a) We illustrate two excited He atoms roaming on the surface of a He droplet following their excitation in the droplet by an XUV pulse from high-harmonic generation sources. Their motion was simulated by a classical trajectory calculation. Atoms 1 and and 2 are initialized at random positions marked by the corresponding symbols and their velocities randomly generated from a Maxwell-Boltzmann distribution at 0.37 K; the weaker He-He interaction are neglected retaining on only the stronger He*-He* interactions. Colored symbols are trajectory endpoints. Both particles propagate until they reach a short interatomic distance where they mutually attract each other and decay via ICD. (b) We plot the mean roaming time τ of He atoms before they meet and decay by ICD as a function of droplet radius R; each value is the result of averaging over Monte-Carlo trajectories. Colors show different initial velocity distributions given by the indicated temperatures. Fitting the simulated curves to a simple power-law for the case of T = 0.37 K reveals that the decay time constant scales for He*-He* ICD in the droplet scales as $\tau \sim R^{3/2}$.*

References:

1) Keshav Sishodia, Ltaief Ben Ltaief, Niklas Scheel, István B. Földes, Andreas Hult Roos, Martin Albrecht, Matyáš Staněk, Lucie Jurkovičová, Ondrej Hort, Jaroslav Nejdl, Ernesto García-Alfonso, Nadine Halberstadt, Jakob Andreasson, Eva Klimešová, Maria Krikunova, **Sivarama Krishnan**, Andreas Heidenreich & Marcel Mudrich XUV fluorescence as a probe of interatomic coulombic decay of resonantly excited He nanodroplets. *Sci Rep* **16**, 1321 (2026).

https://doi.org/10.1038/s41598-025-34677-x
2) Ben Ltaief, L. et al. Efficient indirect interatomic coulombic decay induced by photoelectron impact excitation in large pure helium nanodroplets. *Phys. Rev. Lett.***131**, 023001. https://doi.org/10.1103/PhysRevLett.131.023001 (2023).
3) LaForge, A. C., Ben Ltaief, L., Krishnan, S. R., Sisourat, N. & Mudrich, M. Interatomic and intermolecular decay processes in quantum fluid clusters. *Rep. Prog. Phys.***87**, 126402. https://doi.org/10.1088/1361-6633/ad8fbb (2024).
4) Jahnke, T. et al. Interatomic and intermolecular coulombic decay. *Chem. Rev.***120**, 11295–11369. https://doi.org/10.1021/acs.chemrev.0c00106 (2020).
5) Cederbaum, L. S., Zobeley, J. & Tarantelli, F. Giant intermolecular decay and fragmentation of clusters. *Phys. Rev. Lett.***79**, 4778–4781. https://doi.org/10.1103/PhysRevLett.79.4778 (1997).
6) Krishnan, S. R. et al. Dopant-induced ignition of helium nanodroplets in intense few-cycle laser pulses. *Phys. Rev. Lett.*107, 173402. https://doi.org/10.1103/PhysRevLett.107.173402 (2011)

Posters

Improving phase sensitivity of SU(1,1) interferometer using even and odd coherent states

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We investigate the phase sensitivity of an SU(1,1) interferometer by using nonclassical states. Specifically, both even and odd coherent states, as well as the squeezed vacuum state, are used as inputs. The interferometer setup includes two parametric optical amplifiers and a phase shifter. Intensity detection at the output port is used to measure phase estimation precision via the error propagation method. To quantify the ultimate bounds of phase estimation, we calculate the quantum Cram´er–Rao bound using the quantum Fisher information technique. Our results show that the odd coherent state significantly enhances phase sensitivity compared to the even coherent state and the standard coherent state, this surpasses the standard quantum limit and approaches the Heisenberg limit in certain squeezing parameters. Furthermore, with a larger squeezing parameter, all schemes converge toward a similar phase sensitivity, consistent with the quantum Cram´er–Rao bound predictions.

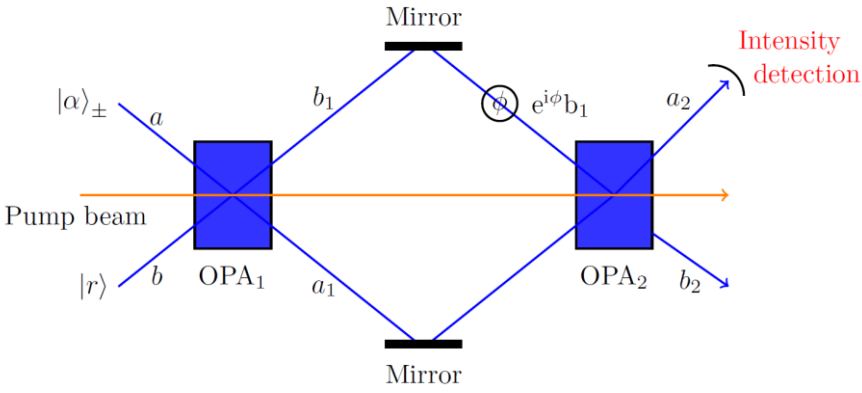

Figure 15: Schematic diagram of the SU(1,1) interferometer.

X-ray Photoelectron Spectroscopy of Doped Helium Nanodroplets with a Hemispherical Electron Analyzer

Narcis-Silviu Blaj[1], Niklas Scheel[2], Rajni Rajni[1], Asbjørn Ø. Laegdsmand[1], Keshav Sishodia[3], Aleksandar Milosavljevic[4], John Bozek[4] and Marcel Mudrich[1,2]

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Helium nanodroplets (HNDs) are superfluid quantum clusters that provide an ultracold and weakly interacting environment, making them ideal inert cryogenic matrices for spectroscopy of embedded atoms, molecules, and clusters [1]. Here we present the first high-resolution X-ray Photoelectron Spectroscopy (XPS) measurements of pure and doped helium nanodroplets, carried out at the PLEIADES beamline of SOLEIL Synchrotron using a hemispherical electron analyzer (HEA). We investigated He droplets doped with krypton and argon. The formation of Ar and Kr clusters was identified through the appearance of additional lower-binding-energy features (see Fig. 1) accompanying the atomic photolines (Kr 3d and Ar 2p, split in spin–orbit doublets of the respective ion). These spectra resemble spectra of pure rare-gas clusters measured earlier [2,3], indicating only weak perturbations by the He droplets.

We systematically recorded XPS spectra as a function of photon energy, droplet size, and doping level and followed the evolution of spectral line shifts and the relative peak intensities of atomic and cluster correlated peaks. The high energy resolution of the HEA allows for a clear separation of overlapping spectral features.

HeDopant simulations [4] were used to estimate the mean dopant number per droplet and to provide qualitative insight into pickup process and helium evaporation. Future experiments at SOLEIL will extend this work to NEXAFS studies of selected dopants in He nanodroplets.

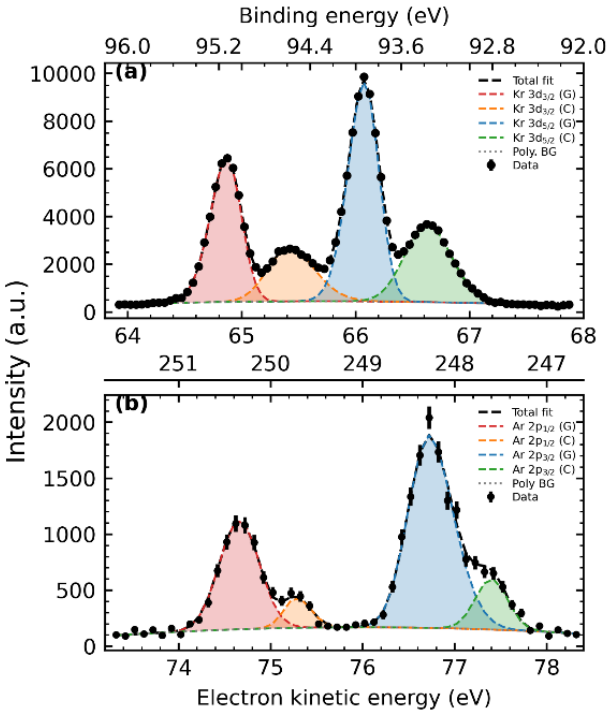

Figure 17: X-ray photoelectron spectra of Kr and Ar clusters. (a) Kr 3d spectrum recorded at a photon energy of hν = 160 eV. (b) Ar 2p spectrum recorded at hν = 325 eV.

References:

1. J. P. Toennies et al., Angew. Chem. Int. Ed. (2004) 43, 2622 – 2648.
2. T. Hatsui et al., J. Chem. Phys. (2005) 123, 154304.
3. M. Tchaplyguine et al., Chem. Phys. (2003) 289, 3.
4. K. Sishodia, S. De, HeDopant: Program to simulate doping processes in helium nanodroplets, (2026) version 1.0.0.

Ultrafast Population Dynamics of Metastable States in Helium Nanodroplets

Shuxing Wang[1], Silviu Blaj[1], Carlo Callegari[2], Michele Di Fraia[3], Andreas Hans[1], Ltaief Ben Ltaief[4], Sitanath Mondal[5], Asbjørn Ørnemark[1], Oksana Plekan[2], Sivarama Krishnan[6], Niklas Scheel[4], Constant Schouder[7], Frank Stienkemeier[5], and Marcel Mudrich[1]

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Here, we investigate delayed low-energy electron emission in He nanodroplets using a pump-probe depletion scheme at the FERMI free-electron laser (FEL). The droplets were ionized by an intense XUV pump pulse, and the resulting electrons and ions were detected by a magnetic-bottle electron spectrometer and an ion time-of-flight mass spectrometer, respectively. A delayed UV probe pulse (~393 nm) partially depleted the long-lived excited-state population, producing pronounced changes in the low-energy electron spectra. These delay-dependent features are observed in a wide range of pump FEL energies (28-68 eV) above the ionization threshold of atomic helium, develop on a timescale of tens of picoseconds, and become dominant at longer delays >20 ps. The probe-induced spectral response includes near-zero-energy electrons and two distinct features converging toward ~1.4 and ~2.2 eV, consistent with ionization of long-lived excited states formed during post-ionization relaxation in the droplet. Contributions from metastable 1*s*2*s* states ionized via a two-photon UV process are discussed as a possible origin of the observed signal.

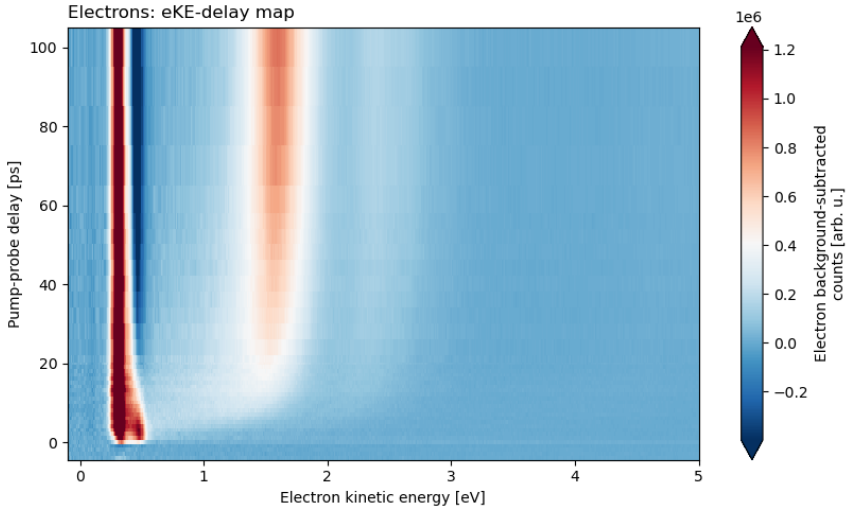

Figure 16: Preliminary pump-probe photoelectron map of He nanodroplets measured at an XUV photon energy of 38 eV.

References:

1. A. C. LaForge, L. Ben Ltaief, S. R. Krishnan *et al*., Rep. Prog. Phys. 87 (2024) 126402.
2. A. C. LaForge, R. Michiels, Y. Ovcharenko *et al*., Phys. Rev. X 11, 021011 (2021)

Experimental and Computational Investigation of Helium Solvated O_3-$(N_2)_N$ Complexes

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The microsolvation of ozone with molecular nitrogen has been studied using helium nanodroplet isolation spectroscopy and quantum chemical calculations. Infrared spectra of O_3 solvated by up to five N_2 molecules were recorded in the region of the asymmetric stretching fundamental, revealing a progression of bands that shift systematically with increasing N_2 pick-up pressure. The O_3-N_2 dimer shows well resolved rotational structure consistent with a T-shaped geometry, in line with previous gas-phase microwave results [1]. Calculations at various levels of theory, including wavefunction-based and density functional theory methods were used to predict vibrational shifts and provide structural insight. The theoretical frequency shift for the dimer is blue-shifted relative to the monomer, consistent with the experiment. Across all levels of theory, RI-CCSD(T)/aug-cc-pVTZ yields good prediction of asymmetric stretch for both O_3 and O_3-N_2 at 1039.25 cm^{-1} and 1041.02 cm^{-1}, respectively. Preliminary results will be presented on identifying the predominant interaction involved in the O_3-$(N_2)_N$ complex.

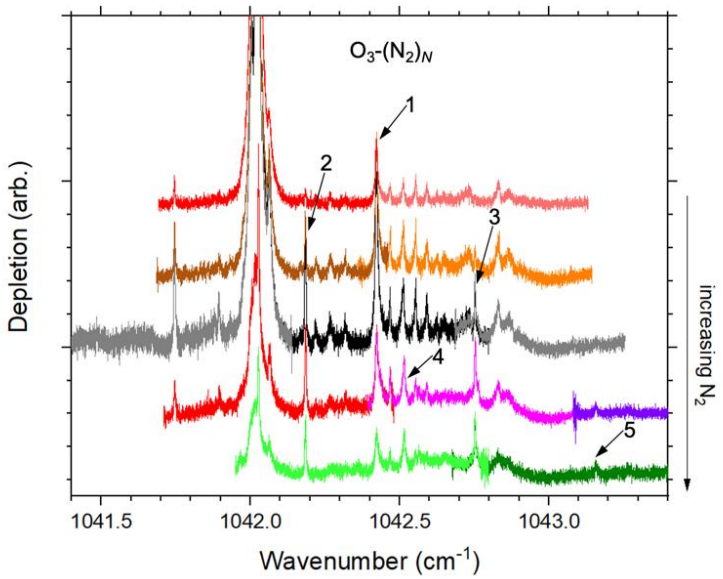

Figure 17: Infrared spectra of O_3-$(N_2)_N$ cluster sizes in helium nanodroplets, where increasing N_2 is labelled from top to bottom.

***References*:**

1. J. P. Connelly, B. J. Howard, *J. Mol. Spectrosc*. (2000), **199**, 205−216.

Modification of spectral properties of PAH cations upon cluster formation

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Polycyclic aromatic hydrocarbons (PAHs) are widely considered key carriers of the unidentified infrared bands (UIBs) observed in the interstellar medium. While the spectroscopic properties of isolated PAHs have been extensively studied, those of ionized PAH clusters remain poorly constrained. In this work, we investigate how cluster formation modifies the spectral properties of PAH cations using helium-tagging action spectroscopy, supported by quantum chemical calculations.

Our results show that clustering significantly alters PAH spectra. In the CH-stretching region, the intensity of the 3.3 µm band is enhanced by more than an order of magnitude while its position remains largely unchanged, confirming its robustness as a tracer of both isolated and clustered PAHs. In the mid-infrared range, clustering leads to band broadening, intensity enhancement, and spectral shifts, particularly in the 7–9 µm region, improving agreement with astronomical observations. In contrast, bands near 6.2 µm remain redshifted, indicating that small PAH cations and their clusters are unlikely carriers of this feature.

Furthermore, cluster formation induces strong broad electronic absorption extending into the infrared, potentially contributing to the observed continuum emission, while absorption in the UV–Vis–NIR range is significantly reduced. These findings suggest that ionized PAH clusters have distinct spectroscopic signatures that must be considered in astrophysical models.

Helium-Tagging Spectroscopy of PAH and Fullerene Cations

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Helium-tagging spectroscopy has emerged as a sensitive method for probing the intrinsic optical properties of cold molecular ions and ionic clusters under well-controlled conditions, while introducing only minimal perturbations to the investigated species. In our approach, ions are generated and efficiently cooled inside multiply charged helium nanodroplets. Controlled collisional heating of the droplets with helium buffer gas enables the controlled formation of helium-solvated ions and ionic complexes at low temperatures. Upon mass selection, resonant photoexcitation of the target ions leads to the evaporation of one or more weakly bound helium atoms. The resulting helium loss is monitored by time-of-flight mass spectrometry and provides an action spectroscopy scheme for recording electronic and vibrational spectra [1]. This technique allows the production and spectroscopic investigation of a broad range of species, including isolated molecular ions, weakly bound ionic complexes, and size-selected ionic clusters. In addition, the implementation of a second ion source makes it possible to generate multiply charged ions, extending the accessible systems also to higher charge states [2].

In this contribution, we focus on the spectroscopy of polycyclic aromatic hydrocarbons (PAHs) and fullerenes in both the visible and infrared spectral regions [3,4]. These carbonaceous molecules are of fundamental interest in molecular physics, materials science, and especially in

astrochemistry, as they are considered potential carriers of diffuse interstellar bands and infrared emission features observed in space. We present spectra of PAH and fullerene cations, providing insight into their electronic transitions, vibrational fingerprints.

***References*:**

1. Bergmeister et al., *Rev. Sci. Instrum.* **94**, 055105 (2023)
2. Ganner et al., *Phys. Rev. Lett.* **133**, 023001 (2024)
3. Ganner et al., *ACS Earth Space Chem.* **9**, 11, 2694 (2025)
4. Ganner et al., *Astrophys. J.* **993**, 47 (2025)

Toward protein matter-wave interferometry: current challenges and the promise of helium nanodroplets

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Demonstrating quantum wave behavior of structurally complex molecules remains long-standing goal at the quantum-classical boundary. Over the past decades, matter-wave interferometry has been demonstrated from diffraction of fullerenes[1] to near-field interferometry with polypeptides[2] and massive nanoclusters exceeding 170 kDa[3]. Especially, near-field effect enables the observation of high-contrast interference fringes even for de Broglie wavelengths of down to 10 femtometers.

Despite the great success, it has remained challenging to achieve this feat also for neutral proteins, which represents a novel material class in these studies. This research is confronted with several outstanding challenges: generating a well-defined, low-velocity molecular beam from complex biomolecules, achieving beam depletion by laser light to realize diffraction gratings, soft ionization for mass detection and detecting and analyzing a necessarily weak beam flux. We have been addressing each of these bottlenecks and recent work demonstrates that photo-cleavable tags attached to biomolecules can be cleaved off in the gas phase by a single photon, and even by visible light[4].

In this presentation, we will discuss the challenges for protein beam experiment and promises of photo-cleavage in the gas phase as well as alternative detection schemes using superconducting nanowires[5], which can bypass ionization entirely. Furthermore, a key open question, the preparation of intense, slow and forward directed beams, will be discussed, which directly connects this talk to the key topic of the conference, i.e. the applications of helium nanodroplet beams.

References

1. M. Arndt, O. Nairz, J. Voss-Andreae, C. Keller, G. van der Zouw, A. Zeilinger, *Nature* (1999), **401**, 680-682
2. A. Shayeghi, P. Rieser, G. Richter, U. Sezer, J.H. Rodewald, P. Geyer, T.J. Martinez, and M. Arndt, *Nat. Comm.* (2020), **11**, 144
3. S. Pedalino, B. E. Ramírez-Galindo, R. Ferstl, K. Hornberger, M. Arndt and S. Gerlich, *Nature* (2026), **649**, 866-870.
4. O. Rybakova, J. Reisinger, P. Rieser, P. Geyer, S. Gerlich, M. Arndt, A. Kumar, D. Häussinger, M. Mayor and V. Köhler, *Helv. Chim. Acta* (2025), e202500022
5. M. Strauß, A. Shayeghi, M. Mauser, P. Geyer, T. Kostersitz, J. Salapa, O. Dobrovolskiy, S. Daly, J. Commandeur, Y. Hua, V. Köhler, M. Mayor, J. Benserhir, C. Bruschini, E. Charbon, M. Castaneda, M. Gevers, R. Gourgues, N. Kalhor, A. Fognini and M. Arndt, *Sci. Adv.* (2023), **9**, eadj2801

Femtosceond time-resolved charge transfer dynamics in chromophore-solvent clusters inside He nanodroplets

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This project uses helium nanodroplets as a cryogenic host to assemble size selected chromophore-solvent clusters and investigates their intermolecular charge transfer dynamics with femtosecond time-resolved photoionization spectroscopy. The main goal is to follow proton-coupled electron transfer (PCET) in isolated molecular aggregates on their natural timescales and to compare the dynamics in helium droplets with the corresponding gas-phase behaviour. The experimental strategy combines correlated electron and ion detection to access both electronic and nuclear motion. Time-resolved photoelectron spectroscopy will be used to track the evolution of the electronic structure and charge redistribution, while ion momentum imaging will provide structural and reaction-coordinate information through photofragment translational spectroscopy (PTS) and Coulomb explosion imaging (CEI). In particular, the velocity map imaging spectrometer combined with a Timepix 3 detector enables cluster-size-selective 3D ion momentum reconstruction and allows the separation of overlapping cluster fragments within the droplet beam. Together, these observables provide a direct view of coupled electron and nuclear dynamics during ultrafast reaction pathways. The project focuses on two complementary classes of PCET reactions. In Branch A, chromophore-to-solvent PCET is investigated in phenol–ammonia clusters ($PhOH – (NH_3)_n$), a benchmark system for which the gas-phase dynamics are already well understood; this branch will establish how helium droplet confinement modifies the known reaction pathways, branching ratios, and fragmentation behaviour. In Branch B, solvent-to-chromophore PCET is studied in pyridine–water clusters ($Py – (H_2O)_n$), where theory predicts transfer from the solvent to the photoexcited chromophore, but time-resolved experimental evidence is still missing. This second branch will provide the first direct measurement of solvent-

to-chromophore PCET in the time domain and determine how the process depends on cluster size and helium-mediated stabilization. By combining helium-droplet assembly, femtosecond pump-probe ionization, electron-ion correlation, and momentum-resolved fragment detection, the project will deliver a mechanistic picture of PCET in finite molecular aggregates. More broadly, it shall establish a general experimental platform for studying ultrafast chemical dynamics in microsolvated clusters with controlled size and environment.

Exploring vortex formation in grazing collisions between superfluid helium nanodroplets and TiO_2

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Superfluid helium nanodroplets (HNDs) exhibit unique properties that have proven advantageous across multiple research areas, including ion spectroscopy, surface deposition, cold chemistry, solvation phenomena, and collision dynamics. Below the critical temperature of 2.17 K, ^{4}He enters the superfluid phase, behaving as a quantum fluid, storing angular momentum through the formation of quantized vortices. While vortices in bulk helium have been studied for decades, early evidence of vortex formation in HNDs was obtained ex situ through the deposition of nanometer-sized filaments, arising from metal atoms binding to vortex cores. More recently, X-ray diffraction imaging has enabled in situ visualization of helium nanodroplets and their embedded species, providing direct insight into droplet shapes and vortex structures.[1]
Theoretically, helium density functional theory (^{4}He-DFT) has been the main ab initio approach to describe HND structure and vorticity.[2] Recent ^{4}He-DFT studies have extended to collisions between pristine HNDs, demonstrating that such interactions can induce the nucleation of quantized vortices. The number and configuration of vortices were found to depend sensitively on parameters such as impact parameter, relative velocity, and droplet size.[3]

Building on this, the present work proposes an alternative scenario in which a single helium nanodroplet is shot towards a stationary TiO_2 surface under grazing incidence. The interaction with the surface is expected to transfer angular momentum, leading to deformed droplets with arrays of quantized vortices within. Ongoing ^{4}He-DFT simulations explore the influence of droplet velocity, droplet size, and incidence angle on this process. In parallel, a recently funded project by the Austrian Science Fund (FWF) aims to investigate the same mechanism experimentally using micrometer-sized droplets, enabling direct visualization via shadowgraphy.

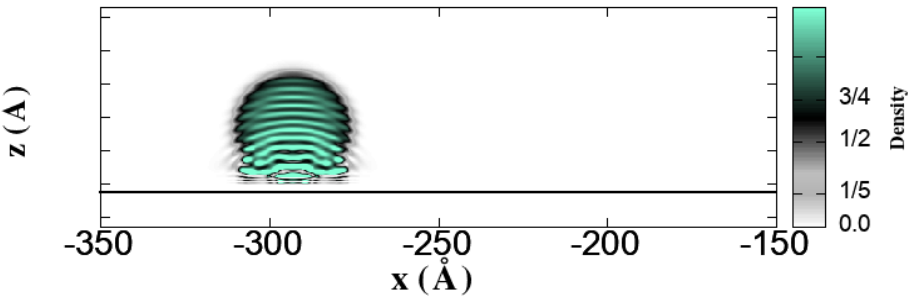

Figure 18: Collision of a 500 He atom droplet with a TiO2 surface at an incidence angle of 85° and an initial velocity of 200m/s.

***References*:**
1. O. Gessner, A.F. Vilesov, *Annu. Rev. Phys. Chem.,* 70, 173-198 (2019)
2. F. Ancilotto *et al., Int. Rev. Phys. Chem.,* 36, 621-707 (2017)
3. E. García-Alfonso *et al., J. Chem. Phys.,* 159, 074305 (2023)

Penning ionization electron spectroscopy of Na and Mg doped in helium nanodroplets

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We investigate the direct photoionization and Penning ionization of alkali (Na) and alkaline-earth metals (Mg) attached to superfluid helium nanodroplets under irradiation with extreme ultraviolet (XUV) synchrotron radiation that resonantly excites the He droplets. Helium is a well-suited system for studying indirect ionization processes due to the simple electronic structure of He, well-resolved electron spectra, weak interaction with embedded species, efficient pick up of molecules and controlled aggregation inside the nanodroplets.
We measured spectra of all electrons and of electrons recorded in coincidence with specific ions for He nanodroplets doped with Mg and Na atoms. For Na, the Penning ionization electron spectrum is well resolved but shifted due to attractive interaction of the Na atom and the excited He* (short-range interatomic Coulombic decay). For Mg, we observe a pronounced enhancement of shake-up ionization where an excited Mg ion

is produced in the Penning process. Shake-up states are only weakly present in direct photoelectron spectra, see Figure.

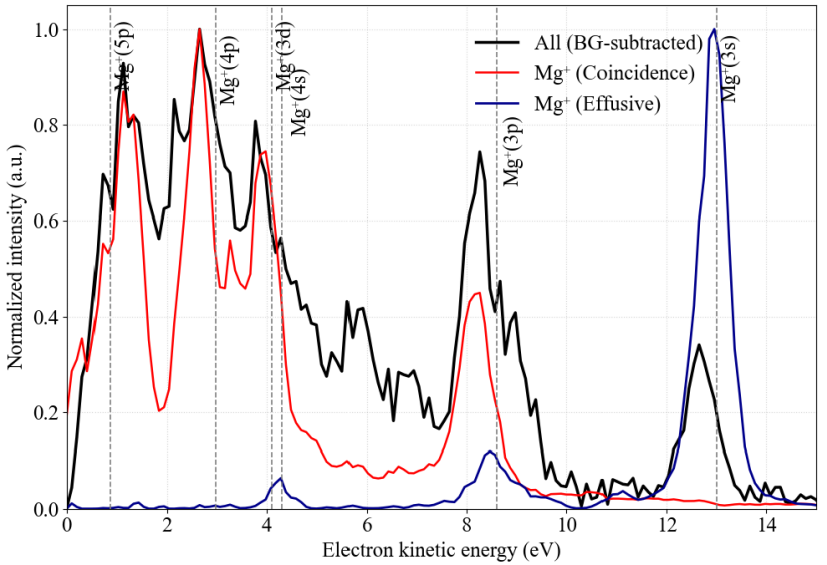

Figure 19: Normalized electron kinetic energy spectra of Mg in helium nanodroplets

Measuring the speed of interatomic Coulombic decay in helium nanodroplets

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Helium nanodroplets are an interesting “test tube” for many types of interatomic reactions. In this work we have looked at both interatomic Coulombic decay (ICD)[1] and laser-assisted electron scattering (LAES)[2]. We have measured the rate of ICD in lithium-doped helium nanodroplets by an EUV-pump NIR-probe scheme. The nanodroplets are resonantly excited into the $2s2p^1P$ state with a 21.6 eV pump pulse and multi-photon ionized with a 1.56 eV probe pulse. ICD and LAES electrons are detected with a magnetic bottle electron spectrometer. Arrival of the probe pulse ionizes most of the $1s2p^1P$ He, quenching the ICD process. By varying the time delay between pump and probe we measure the speed of the ICD for different droplet sizes, as shown in Fig 1. From the resulting electron spectra we distinguish several ionization pathways, such as ICD, LAES and above threshold ionization(ATI). In contrast to previous measurements, where ICD takes place in a matter of picoseconds[3], in our large droplets, the measured lifetime of the ICD process extends into the nanosecond range. We propose a model for the dynamics where excited states travel to the surface and roam until they meet another excited state. When these surface states are ionized the process is through LAES. They can also desorb from the droplet surface, in which case they will ionize through ATI.

Using this model we determine the rate constants of both the ICD process and the desorption of excited helium atoms from the nanodroplets

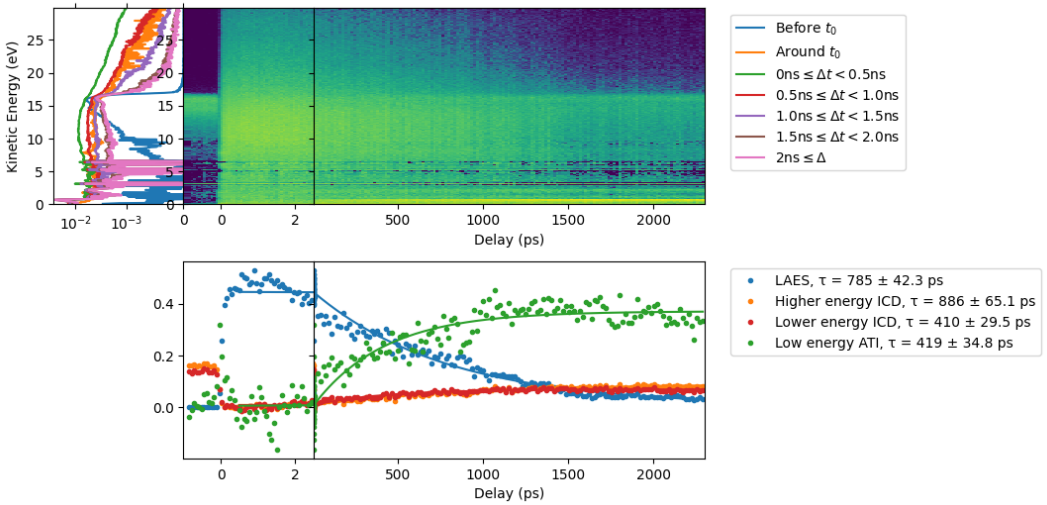

Figure 20: The dynamics of ICD in very large helium nanodroplets

References:
1. L. Ben Ltaief, Phys. Rev. Res. 6, 013019 (2024).
2. L. Treiber, Nat Commun 12, 4204 (2021).
3. A. C. LaForge, Phys. Rev. X 11, 021011 (2021).

Interaction dynamics between organic dopants and a helium nanodroplet environment probed with time-resolved photoelectron spectroscopy

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Time-resolved photoelectron spectroscopy of single organic molecules embedded in helium nanodroplets (HNDs) allows for the study of intramolecular relaxation dynamics with the advantage of a cold and weakly perturbing environment. For the unambiguous interpretation of the data it is inevitable to characterize the helium environment's influence on the electrons emitted from the dopant, as it has been reported to significantly impact the measured photoelectron signal [1]. Moreover, it is essential to understand the HND-dopant interaction, to account for its impact on the investigated dynamics. In this work, we study the interaction and photoinduced dynamics of tetracene molecules embedded in HNDs. We observe a droplet size-dependent influence on the photoelectron spectrum by the surrounding helium. Furthermore, we identify signatures in the time evolution of low kinetic energy photoelectrons that indicate a complex interplay between two processes upon photoexcitation of the organic dopant: helium evaporation and dopant ejection (see figure 1). Both phenomena are well-known independently. Helium evaporation for instance is commonly used for the investigation of non-radiative transitions by depletion spectroscopy [2] and the ejection of dopants has been observed before for In atoms [3] and In_2 molecules in HNDs [4, 5]. A systematic study of the helium-dopant dynamics triggered upon photoexcitation of the tetracene dopants will be presented.

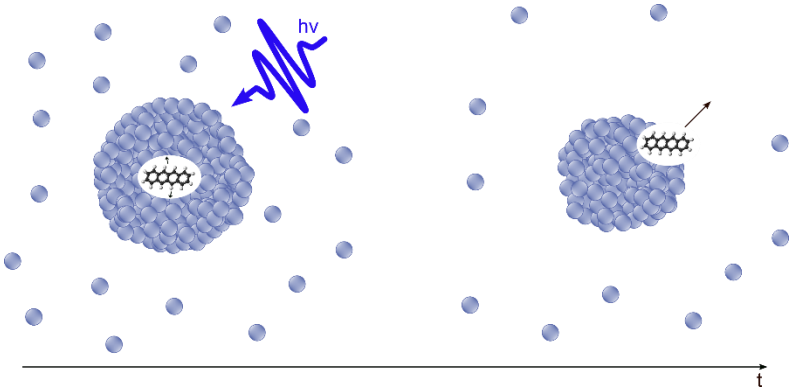

Figure 21: Helium evaporation and dopant ejection upon photoexcitation

***References*:**

1. Loginov, E. et al. (2005), *Phys. Rev. Lett.* **95** (16): 163401
2. Toennies, J.P. and Vilesov. A. F. (2004), *Angew. Chem. In. Ed.* **43** (20): 2622-2648
3. Thaler, B. et al. (2018), *Nat. Commun.* **9** (1): 4006
4. Meyer, M. et al. (2019), *EPJ Web Conf.* **205**: 06005
5. Thaler, B. et al. (2020), *Phys.Rev. Lett.* **124**: 115301

Ultrafast dynamics in photoexcited Helium nanodroplets using compact high-harmonic source at ELI Beamlines facility

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Ultracold He-nanodroplets form a unique spectroscopic environment for studies of photochemical processes involving dopant atoms, molecules and small aggregates. The combination of a compact high-harmonic (HHG) extreme ultraviolet (XUV) source with advanced spectroscopic techniques provides access to the intra- and interatomic energy re-distribution dynamics in photoexcited He nanodroplets. In this contribution recent achievements and feature developments driven by the He-nanodroplets user community at the user end-station MAC will be highlighted [1-3].

The MAC end-station at the ELI Beamlines facility [4-6] is available via open user calls. The end-station is equipped with detectors to measure fluorescence photons [3], charged particles via velocity map imaging,

magnetic bottle type spectrometer [5] or with 3D momentum imaging based on the TPX3CAM technology [6]. The generation of circularly polarized XUV pulses and ultra-short pulses in the deep ultraviolet spectral region are under development.

Resonant XUV excitation of individual atoms inside the He nanodroplet [2] initiates complex electronic and nuclear relaxation dynamics. Depending on the chosen parameters of the temporally delayed NIR-probe pulse different pathways can be selectively probed. Intense NIR pulses initiate avalanche ionization and nanoplasma ignition in XUV excited nanodroplets. This way, the formation of so called “snowball” complexes [1] and roaming of He* atoms at the droplet surface [3] is observed. At NIR pulse intensity below plasma ignition threshold laser-assisted electron scattering is observed [A. Lægdsmand, *in preparation*].

***References*:**

1. C. Medina *et al*., *New J. Phys.* (2023), **25**, 053030
2. L. Jurkovičová ***et al.***, *Commun. Phys.* (2024), **7**, 26
3. K. Sishodia *et al*., *Sci. Rep.* (2026), **16**, 1321
4. E. Klimešová *et al.*, *Eur. Phys. J. Spec. Top.* (2021), **230**, 4183
5. E. Klimešová *et al*., *Acta Phys. Pol.* A (2024), **145**, 118
6. D. Ševaev *et al., JINST* (2026), **21,** C03022

Correlated electron and ion imaging of near-infrared induced nanoplasma in helium nanodroplets

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We report single-shot correlated measurements of electrons and ions emitted from doped helium nanodroplets exposed to intense (~10^{15} Wcm^{-2}) near-infrared (NIR) laser pulses. By combining electron velocity map imaging (VMI) with ion time-of-flight (TOF) spectroscopy, we simultaneously obtain electron momentum distributions and ion TOF spectra on a shot-by-shot basis. This approach allows direct correlation of electron and ion kinetic energies originating from individual nanoplasma events. We use a novel approach that utilizes the ion TOF spectrometer to determine the kinetic energies of ions emitted after nanoplasma explosion. Analysis of the average charge of emitted ions reveals two distinct nanoplasma populations: one primarily dominated by He^+ ions and the other by He^{2+} ions, see figure (1b). These two regimes exhibit significantly different electron and ion kinetic energy distributions. Electrons correlated with He+-dominated nanoplasma events display low kinetic energies (< 1 eV), which we attribute to the ionization of Rydberg atoms by the electric field of the VMI spectrometer [1,2], consistent with earlier single-shot studies [3]. In contrast, the electrons correlated with He^{2+} ions show a shift in emitted kinetic energy, see figure (1c). The correlated ions possess kinetic energies of several eV, arising from Coulomb explosion within the charged nanoplasma [4,5]. Overall, these results provide direct insight into the formation and expansion dynamics of nanoplasma.

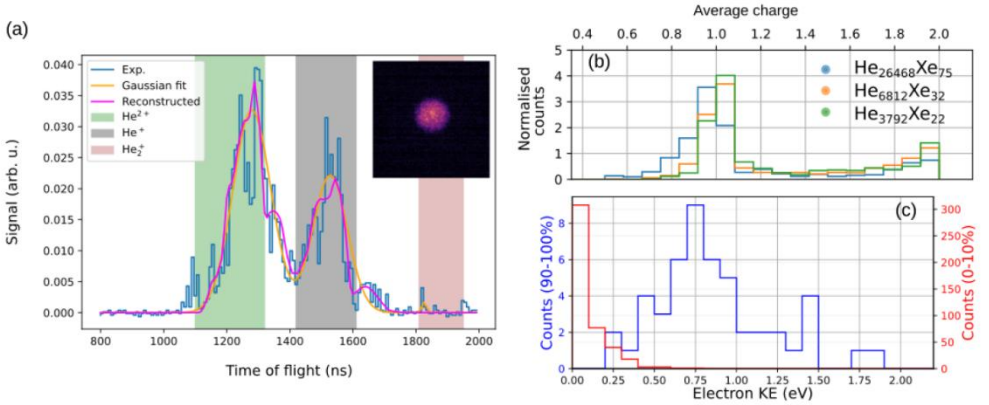

Figure 22:(a) Single shot correlated ion tof spectrum with electron VMI as an insert. (b) Histogram of average charge of emitted nanoplama events shows a 'U'-shaped distribution peaking at average charge of 1.0 (He+) and average charge of 2.0 (He2+). (c) Histogram of electron kinetic energy. Blue curve shows the histogram for the nanoplama events dominated by He2+ (> 90%), and red curve shows events dominated by other ions (< 10%).

References:

1. B. Schütte et. al. New J. Phys. 17, 033043, (2015).
2. D. Komar et. al. Phys. Rev. Lett. 120, 133207 (2018).
3. C. Medina et. al., New J. Phys. 23, 053011 (2021).
4. Md. R. Islam et. al., Phys. Rev. A 73, 041201 (2006).
5. A. Mikaberidze et. al., Phys. Rev. Lett. 102, 128102 (2009).

Investigation of Quinacridone and its microsolvation complexes by Helium Nanodroplet Isolation Spectroscopy

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Organic chromophores are of central importance in applications such as organic solar cells and organic light-emitting diodes (OLEDs), and are emerging candidates for photocatalytic water splitting [1]. A particularly interesting example is quinacridone, widely used in printing pigments, exhibiting complex aggregation behavior. While achiral in isolation, aggregation on surfaces can lead to the formation of chiral structures [2]. Quinacridone and its derivates have also been proposed for photocatalytic water splitting [3]. Understanding its vibronic level structure and relaxation dynamics and how they are affected by the presence of water is a prerequisite for understanding its potential photochemical reactivity.

Helium nanodroplet isolation spectroscopy provides a suitable approach for investigating cold molecules with low vapor pressures. The ultracold, inert helium environment enables probing excited states from the vibrational ground state. In combination with tunable nanosecond laser sources, fine details in the vibrational structure can be investigated.

The complex structure of the observed excitation spectra indicates possible contributions of various tautomers. By varying the doping conditions in helium nanodroplets, different aggregation states, including isolated molecules in different tautomeric forms, dimers, and larger oligomers, can be formed and systematically studied. Toward photocatalytic water splitting, measurements of chromophore–water complexes have been performed and indicate reduced fluorescence yields. The potential formation of dark states and non-radiative relaxation

pathways in these complexes will be investigated with alternative detection schemes based on the photoionization of excited molecular complexes.

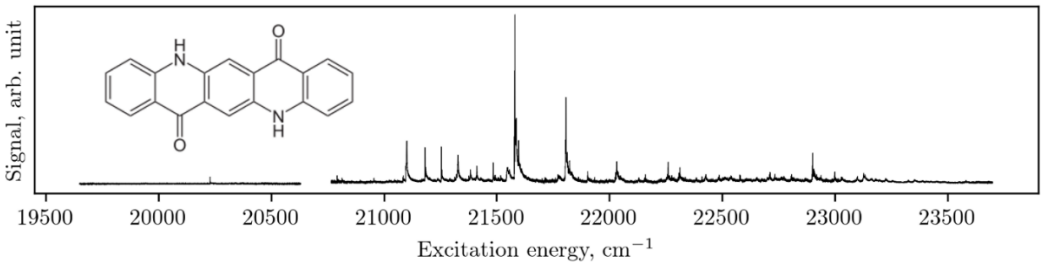

Figure 23: Structure and excitation spectrum of quinacridone.

***References*:**

[1] X. Liu, T. N. V. Karsili, A. L. Sobolewski, & W. Domcke, *Chemical Physics* (2016), 464, 78–85.
[2] N. Humberg, R. Bretel, A. Eslam, E. Le Moal, & M. Sokolowski, *The Journal of Physical Chemistry C* (2020), 124(45), 24861–24873.
[3] E. Rossin, Y. Yang, M. Chirico, G. Rossi, P. Galloni, & A. Sartorel, *Energy Advances* (2024), 3(8), 1894–1904.

Locating Na-Mg dimers on Helium Nanodroplets: A Laser-Induced Coulomb Explosion Study

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The solvation site of dopants in superfluid helium nanodroplets remains a subject of ongoing debate, particularly for alkaline-earth metals. While alkali metal atoms are known to reside on the droplet surface, alkaline-earth atoms like Ca and Sr have contradictory reports suggesting both surface-bound states and submerged interior solvation.[1,2] Therefore, solvation environment of alkaline-alkaline earth metals heterodimers remains a subject of significant interest.

This study investigates the "tug-of-war" within the Na-Mg heterodimer using laser-induced Coulomb explosion imaging. Here, we ionized our molecule by a femtosecond laser pulse leading to Coulomb explosion into Na^+, Mg^+ and Mg^{2+} fragment ions. The mass and velocity of all ions are recorded by the combination of a VMI spectrometer and a Tpx3CAM detector.[3] Using radial and angular covariance maps, we compare the NaMg heterodimer signal with homonuclear Na and Mg molecule. The helium attachment dynamics provide a clear qualitative indicator of the solvation site: pure Mg doping yields extensive Mg^+He_n solvation chains, characteristic of interior submersion. The NaMg produces small fraction of helium attached fragments. The negligible helium pick-up during fragment escape suggests the heterodimer resides on the droplet surface. However, the angular covariance features for NaMg are found to be narrower than those of surface-bound requiring more precise probe of the molecule.

Further, Ion deflection angles were needed to give quantitative confirmation of the solvation site. Trajectory bending occurs as fragment ions polarize the surrounding helium, inducing an attractive potential causes deflection. We found a mean deflection of 4.8° at 15 K, which increased to 6° as a function of droplet size at 13.5 K. This scaling behaviour is consistent with the surface-bound Na_2 systems and confirm

the location of this heterodimer to be near the surface. Further it would be very interesting to investigate other alkali-alkaline earth combinations, e.g. RbMg or LiCa, to further map the competitive solvation landscape of these complex metal species.

References:

1. F. Stienkemeier, F. Meier, H.O. Lutz,, J. Chem. Phys. 1997,107,10816.
2. A. Hernando, R. Mayol, M. Pi, M. Barranco, F. Ancilotto, O. Bünermann, F. Stienkemeier, J. Phys. Chem. A 2007 *111* (31), 7303-7308.
3. C. Schouder, A. S. Chatterley, M. Johny, F. Hübschmann, A. F Al-Refaie, F. Calvo, J. Küpper, H. Stapelfeldt, Phys. Chem. Chem. Phys. (2021), 23, 11114–11124.

Experimental Solution of the Thomson Problem

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The Thomson problem is a mathematical problem that has been around for more than a century[1]. It addresses the optimal arrangement of *N* like-charges on a conducting sphere. Analytical solutions only exist for a handful of systems containing less than 13 charges. For large *N* the Thomson problem is frequently used as a benchmark for global optimization algorithms. To the best of our knowledge, superfluid helium nanodroplets provide the first experimentally viable platform for approaching the Thomson problem under near-ideal physical conditions: delivering mobile charges on a nearly spherical surface, at ultra-low temperature, with minimal external perturbation.

In multiply charged helium droplets like-charge Coulomb repulsion pushes the charge centers to the surface where they adopt a minimum energy configuration in the form of the famous Thomson problem[1,2]. Coherent diffractive imaging of large multiply charged helium droplets doped with xenon confirms that most for the dopants are found in the form of clusters close to the surface of the droplet[3], however, the quality of the images does not allow a comparison with the solution of the Thomson problem for any specific number of charges.

By doping multiply charged helium droplets with gold vapor, monodisperse singly-charged nanoparticles containing typically several hundred atoms are formed. Upon soft-landing onto a transmission electron microscopy (TEM) grid, each droplet prints its gold particles onto the substrate. The orthographic projection of the numerical solution of the

Thomson problem[4] can be fitted to the experimental TEM images of individual droplets by appropriate rotation and scaling. For droplets with a final radius smaller than 200 nm we obtain excellent agreement between experimental and numerical results. Regular hexagonal patterns with five-fold disclinations observed in imprints of helium droplets with a radius larger than 400 nm indicate that particles located on the back hemisphere are splashed sideways and backwards[5].

***References*:**

1. J. J. Thomson, *Phil. Mag.* (1904), **7**, 237–265.
2. E. Garcia-Alfonso et al., *J. Chem. Phys.* (2024) **161**, 224303.
3. A. J. Feinberg et al., *Phys. Rev. Res.* (2022), **4**, L022063.
4. D. J. Wales, S. Ulker, *Phys. Rev. B* (2006), **74**, 212101.
5. P. Martini et al., Phys. Rev. Lett. (2021), **127**, 263401.

Ultrafast bimolecular reaction catalysis via engineered nuclear quantum effects in helium nanodroplets

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Nuclear quantum effects (NQEs) fundamentally alter chemical reactivities, particularly those occurring at low temperatures or involving light nuclei. Yet, directly resolving how NQEs influence the ultrafast dynamics of bond formation remains a central challenge. Here we report the time-resolved observation of NQEs catalyzing a light-induced bimolecular reaction within a quantum-phase environment. Using femtosecond pump-probe spectroscopy on H_2-H_2 and D_2-D_2 dimers confined in helium nanodroplets at 0.37 K, we find that the low-temperature conditions engineered by the host environment drive a pronounced acceleration of reaction dynamics. The formation of H_3^+ is accelerated to 59 ± 10 fs in the droplet compared to 152 ± 17 fs in the gas phase. This catalytic effect is strongly isotope-dependent, with the heavier D_3^+ exhibiting only a slight reduction in formation time from 153 ± 8 fs to 111 ± 9 fs. Molecular dynamics simulations reveal that the acceleration is catalyzed by pronounced NQEs

under cryogenic confinement, with lighter hydrogen isotopes exhibiting greater quantum delocalization and consequently faster reaction rates. These findings provid a time-resolved perspective on quantum effects in ultracold chemistry, demonstrating how the unique environment of nano-cryo-reactors can harness temperature-driven NQEs to steer the ultrafast dynamics of chemical synthesis.

Localization and Delocalization of a Single Molecule in a Helium Nanodroplet

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Understanding the quantum behavior of molecules in a superfluid environment is crucial for exploring confinement and solvation effects at the nanoscale. Here, the spatial extent of wavefunctions for single molecules embedded in superfluid helium nanodroplets (He_N) is investigated through the analysis of photoelectron momentum distributions (PMDs) from above-threshold ionization [1,2]. By comparing the angular nodal structures in PMDs for light H_2 and heavier D_2 molecules in He_N, distinct localization and delocalization behaviors are identified. The H_2 molecule at a temperature of 0.37 K, with a de Broglie wavelength comparable to the droplet size [3], exhibits delocalization across the droplet, leading to preserved nodal structures in PMDs due to minimal electron scattering. In contrast, the heavier D_2 molecule is more localized within the droplet, resulting in blurred nodal structures as photoelectrons undergo significant scattering with the helium environment. These findings, supported by Monte Carlo and time-dependent Schrödinger equation simulations, reveal how the confining potential of He_N governs the wave-particle duality of solvated molecules, offering new insights into quantum solvation dynamics.

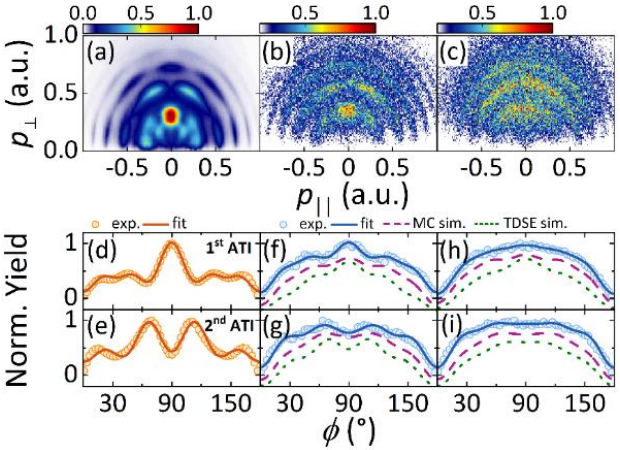

Figure 24: Photoelectron momentum spectra. (a)–(c) Measured PMDs associated with ionic fragments of (a) H+, (b) HeH+, and (c) HeD+ plotted in the ($p_{||}$, $p_{\perp}$) momentum space. (d)–(i) The PADs for the first and second-order ATI associated with (d),(e) H+, (f),(g) HeH^+, and (h),(i) HeD^+.

***References*:**

1. Z. Ye et al., *Phys. Rev. Lett*. (2025), **135**, 213202.
2. T. Wilbois and H. Helm, *Phys. Rev. A*. (2011), **84**, 053407.
3. L. De Broglie, *Compte Rendus*. (1923), **177**, 507.

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