DESIREE Symposium 2022

Monday, 22 August 2022 - Wednesday, 24 August 2022

Albano Building 2

Book of Abstracts

ELISA – the world's first (?!) electrostatic storage ring

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I will give a description of my personal involvement in the conception, development and first years of operation of the ELISA electrostatic storage ring. The realization of such an electrostatic storage ring might in retrospect seem trivial, but at the time many concerns arose.

I believe the success was due to the availability of the right resources and experience at the right time including:
1. The construction and operation of the MAGNETIC dual purpose ion AND electron storage ring ASTRID
2. Knowledge, experience and design tools for storage rings
3. Experiments with atomic and molecular ions in ASTRID
4. Experience with electrostatic elements
5. Mature technical design and workshops

Some "pre-historic" developments will be presented, as will some of the early physics results at ELISA. I will not give an overview of the field, but mention some of the little and bigger "sister" rings and "cousin" traps.

Effect of the anion orbital momentum on mutual neutralization processes

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Recent improvement of the merged beam setups allowed to study the partial cross section for a wide range of atom-atom mutual neutralization (MN) reactions. This range is mainly limited by two factors: The ability to produce the ion beams (ideally in a single electronic state) and the mass ratio between the anion and the cation. This mass ratio limit is around 10 for the UCLouvain single pass setup and 20 at the DESIREE facility.

Therefore, it does not allow studying reactions with higher mass ratios, such as Fe^{+}+H^{-}. To circumvent this experimental limit, it would be useful to replace the H^{-} anion by another anion with similar properties in order to indirectly measure this reaction.

Using the UCLouvain merged beam setup, we recently measured the branching ratio for the MN between C^{+}/N^{+}/O^{+} and C^{-}/Si^{-}/O^{-}/P^{-}/D^{-}/S^{-} [1]. This systematic study allowed us to make a comparative study, e.g., the effect of the anion's binding energy on the outcome of MN reactions. But this analysis in terms of electron affinity was guided by previous studies which mainly focused on reactions involving anions in an *S* state. While the anion binding energy dependency was effectively observed, the branching ratios differ largely for systems whose anion has a similar binding energy but a different orbital momentum, e.g., C^{-}/O^{-} and D^{-}/P^{-}.

Using an anion centered asymptotic model combined with a multi-channel Landau-Zener model, we have found that higher anion orbital momentum increases the number of Λ molecular symmetries in which the collision happens and that the interaction potential between ionic ($A^{+}B^{-}$) and covalent ($A^{0}B^{0}$) states decreases for higher Λ. This allows the production of lower excited states of the neutralized cation, but also lowers the partial cross section for all other states, by distributing the reaction probability among several symmetries.

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Two-body vs three-body processes in mutual neutralisation reactions of atmospheric ions at DESIREE

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In the highest layer of Earth's atmosphere, situated approximately 150 km above the surface, particles are exposed to high levels of radiation and are mostly composed of atomic ions, dominated by O^+ and N^+. During the night, these are neutralised through recombination with free electrons or by interactions with anions [1], a phenomenon known as mutual neutralisation (MN). The process is straightforward to write out:
$A^+ + B^- \rightarrow A^* + B$,
i.e the captured electron ends up in an electronically excited state of the neutralised atom.

In lower layers however, less ionising radiation is present, due to the absorption from the F layer and the higher pressure, and it is believed that the most abundant ions are molecular, i.e N_2^+, NO^+ and O_2^+. In the MN of such diatomic cations with an atomic anion, two different outcomes need to be considered:
$AB^+ + C^- \rightarrow AB+C$
$AB^+ + C^- \rightarrow A+B+C$
i.e, here the captured electron could also end up on a repulsive potential of the formed molecular neutral, such that it fragments, yielding three products.

At the unique cryogenic electrostatic double storage ring DESIREE, these reactions can effectively be studied [3,4]. We present here the first merged beams results of several atmospheric molecular ions, namely O_2^+, NO^+, and N_2^+ interacting with O^-. The results are interesting, as both processes are observed with very different populations depending on the molecular cation, with no dependence on the bond energy. For the three-body processes, very distinct dynamics are observed, with a clear vibrational dependence on the product yield. These results can be partially be explained from a theoretical approach based on dissociative recombination.

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Slow deexcitation dynamics of molecular ions stored in RICE and TMU E-ring

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We have studied isolated molecular ions in a vacuum by taking advantage of two electrostatic ion storage rings, RICE in the cryogenic condition and TMU E-ring at room temperature over the years. We have focused on the deexcitation dynamics of laser-excited carbon clusters (C_n^-) and understood them based on statistical descriptions. This strategy was successfully applied for larger molecules, i.e., pentacene anions ($C_{22}H_{14}^-$) [1].

Recently, we have started rovibrational state-selective observation of the deexcitation of small molecular ions using a wavelength-tunable pulsed laser. The first example is the vibrational cooling of triatomic molecular ions N_2O^+ in the range of several seconds. We found that the deexcitation dynamics drastically deviate from the statistical description based on a Boltzmann-type distribution [2,3]. We also observed deexcitation of the simple iso-nuclear diatomic negative molecule C_2^- in the tens of millisecond range. An iso-nuclear diatomic molecule has zero dipole moment, and the vibration transition is prohibited due to its symmetry. However, it is de-excited via spontaneous infrared radiation between electric excited and ground states. The time evolution of the population at specific vibrational states was measured up to 60 milliseconds, providing the first quantitative experimental support for long-standing theoretical predictions [4]. We also tackled Si_2^- on the micro-second time region. To our surprise, the excitation spectra show well-resolved rovibronic structures exclusively in the delayed time region [5].

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Statistical vibrational autodetachment and radiative cooling rates of *para*-benzoquinone radical anions

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We report measurements of the statistical vibrational autodetachment (VAD, also called thermionic emission) and radiative cooling rates of isolated *para*-benzoquinone (pBQ, $C_6H_4O_2$) radical anions using the cryogenic electrostatic ion storage ring facility DESIREE [1]. The results are interpreted using master equation simulations with rate coefficients calculated using statistical detailed balance theory. The VAD rate is determined by measuring the time-dependent yield of neutral pBQ due to spontaneous electron emission from a highly-excited ensemble of anions formed in an electron-attachment ion source. Competition with radiative cooling quenches the VAD rate after a critical time of $\tau_c = 11.00(5)$ ms. Master equation simulations which reproduce the VAD yield provide an estimate of the initial effective vibrational temperature of the ions of 1100(20) K, and provide insight into the anion formation scenario. A second measurement of the radiative cooling rate of $p\mathrm{BQ}^-$ stored for up to 0.5 s was achieved using time-dependent photodetachment action spectroscopy across the ${}^2A_u \leftarrow {}^2B_{2g}$ and ${}^2B_{2u} \leftarrow {}^2B_{2g}$ transitions. The rate at which hot-band contributions fade from the action spectrum is quantified by non-negative matrix factorisation. This is found to be commensurate with the average vibrational energy extracted from the simulations, with $1/e$ lifetimes of 0.16(3) s and 0.1602(7) s, respectively. Implications for astrochemistry are discussed.

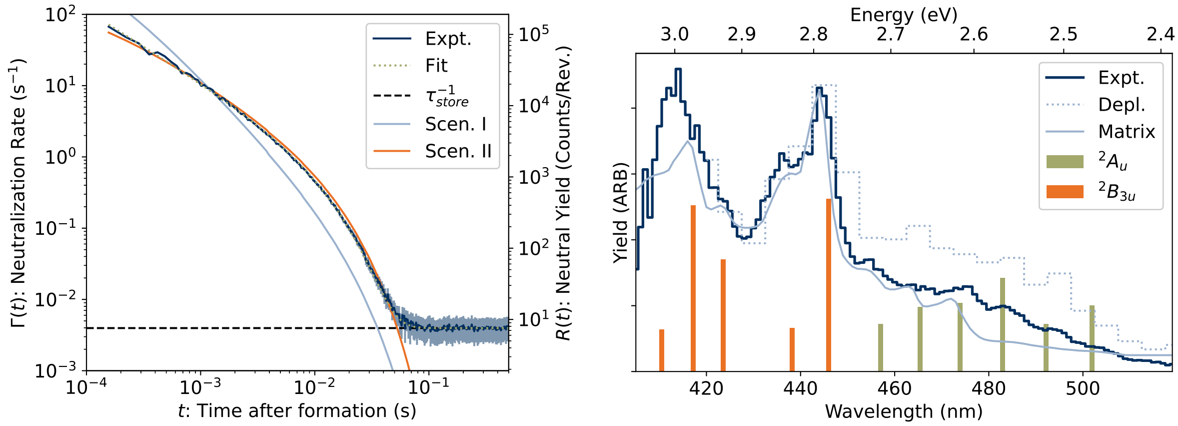

Figure 1: Left: Neutral yield from $p\mathrm{BQ}^-$ formed in an electron attachment ion source. Right: Action spectrum of $p\mathrm{BQ}^-$ integrated over 0.5–5 s storage time.

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Spectroscopy and photoisomerization of protonated Schiff-base retinal derivatives *in vacuo*

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The protonated Schiff-base retinal (RPSB) is responsible for many light triggered reactions in nature. Among them is vision in mammals, where upon absorption of a photon the retinal chromophore undergoes isomerization from its native 11-*cis* configuration to the all-*trans* configuration[1], starting a process which eventually sends a signal to the brain. In the rhodopsin protein in the eye the isomerization occurs within 200 fs[2] with a high quantum yield of 67 %, while in gas phase the isomerization from 11-*cis* to all-*trans* occur within 400 fs and the isomerization from all-*trans* to 11-*cis* occur within 3 ps[3].

To further understand this important biochromophore, we examine how the absorption and dynamic of RPSB changes when steric constraint are introduced to the molecule. Three derivatives of RPSB are examined, including $C_9{=}C_{10}$ *trans*-locked, $C_{13}{=}C_{14}$ *trans*-locked and a retinal derivative without the β-ionone ring.

The absorption in the visual regime was fairly similar for all three derivatives. The major differences in the absorption spectra was found in the UV. Measurements of the excited-state lifetimes showed that by introducing steric constraints, the potential-energy landscape of the chromophore is changed in a way which is not intuitively clear. Locking a single carbon bond does not only correspond to closing of a specific isomerization path with an increased lifetime as a result. Locking the $C_9{=}C_{10}$ bond in the *trans* configuration results in a single decay path out of the first excited state with a lifetime of (717 ± 34) fs. Locking the $C_{13}{=}C_{14}$ bond in the *trans* configuration results in a two-component decay with a short lifetime of (869 ± 98) fs. Note that these lifetimes are both shorter than the 3 ps observed for the unlocked all-*trans* retinal chromophore. Removing the β-ionone ring results in a single exponential decay with a lifetime of (895 ± 59) fs.

Our work shows that by introducing chemical modifications, both the absorption and excited-state dynamics of gas phase protonated Schiff-base retinal may be significantly altered.

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Laser Photodetachment Studies on negative Ions in the Frankfurt low energy Storage Ring

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The low–energy electrostatic storage ring FLSR at the University of Frankfurt [1] is a room temperature storage ring with 14.23 m circumference in a race track geometry. It allows to store heavy ions up to 50 keV kinetic energy. Recently, it has been equipped with a negative ion source to store atomic and molecular negative ions and to study laser photodetachment of vibrationally cooled molecules. Storage of O^- and OH^- as well as the metastable He^- has been achieved [2]. At a target point in the ring with enhanced ion density optical grade vacuum windows have been installed to allow crossed beam interaction with laser photons. A high–repetitive tunable Ti:sapphire laser with 10 kHz rate developed at the University of Mainz allows to perform photodetachment spectroscopy around the detachment threshold. The neutralized particles can be detected further downstream by position sensitive double sided silicon strip detectors, which are mounted at the end of the straight section of the ring. By varying the laser wavelength the detachment threshold of the negative ion and therefore the electron affinity (EA) can be studied. Preliminary studies on O^- gave promising results.

The precise knowledge of the EA of negative ions is important for a recent project in which laser photodetachment is used as an element selective filter to suppress isobars in accelerator mass spectrometry (AMS). For most atomic negative ions the EA is well known. However, in the case of molecules the EA is not always known and the lack of spectroscopic information especially of AMS relevant molecular negative ions is evident. One reason for this are the internal degrees of freedom of molecules. In that case more effort has to be taken in gathering the required spectroscopic information. The study of vibrationally cooled molecules in the ring can help to provide the required data and make new isotopes accessible for AMS.

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Elephants Flying Through Space in a Box: The DESIREE Electrospray Ion Source

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An electrospray ionization (ESI) ion source is currently being commissioned at DESIREE. The source has been designed to produce short, intense bunches of pre-cooled molecular ions for injection into the DESIREE storage rings [1]. The source will expand DESIREE capabilities by allowing introduction of large and/or fragile molecules, such as biochromophores, molecules important in atmospheric sciences, and other carbonaceous systems, e.g. substituted PAHs, which are not accessible with other ion sources.

A central feature of the new ESI source is coupling with a cryogenically cooled ring electrode trap (RET), from which short, intense bunches of internally cold molecular ions are extracted and injected into the DESIREE facility.

In an expert commissioning run, bunched beams of Rhodamine 640 cations were stored in DESIREE after trapping in both a room-temperature octupole pre-trap and the cryogenic RET. In both cases, measurable but small ion currents (10s of picoamps) were detectable at the end of each storage cycle. Ejection electrodes on the octupole pre-trap increase the stored beam current, but also heat some portion of the bunch, leading to spontaneous dissociation, as shown in the left figure below. The right figure shows the laser-induced neutral yield rate following the first few laser shots for room-temperature Rhodamine ions from the pre-trap. No laser-induced signal was observed for cold Rhodamine ions from the RET.

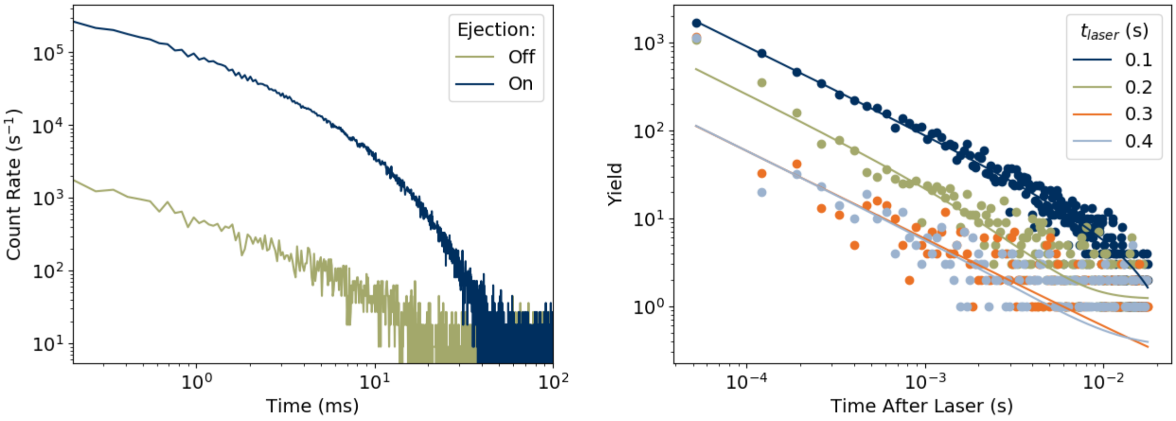

Figure 1: Left: Spontaneous dissociation of Rhodamine 640 cations due to ejection electrodes Right: Laser-induced dissociation of Rhodamine ions.

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Photoionization and fragmentation of cyclic dipeptides, experimental and computational efforts on the search of the 'seeds of life'

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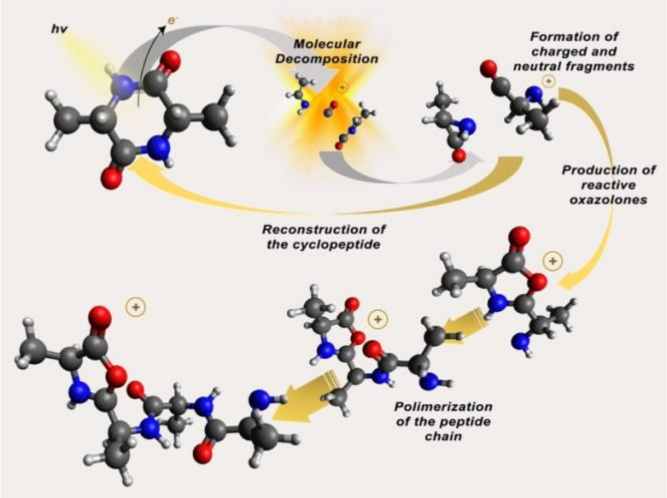
Since the well-known Miller's experiment [1] in the 50s, the origin of biologically relevant compounds in abiotic conditions has attracted a lot of interest. Several investigations on the synthesis of early Earth organic compounds have been reported and the concept of a 'prebiotic soup', where some kind of activation triggers the spontaneous formation of such species, has been proposed as a likely milieu for the first appearance of complex molecules necessary for life.

In this work, we propose a new and alternative synthetic route for the formation of peptide chains that does not require a catalyst or an aqueous environment. In a collaboration among the groups at CNR-ISM, University Autonoma Madrid, Stockholm University and Elettra Sincrotrone Trieste state-of-the-art experiments and extensive simulations have been combined to show that VUV ionizing radiolysis of cyclo(alanine-alanine) [2,3], one of the simplest 2,5-Diketopiperazines (DKP) produces reactive cationic and neutral fragments such as oxazolidinones intermediates. Reactions of these fragments may lead to the reconstruction of the original cyclic dipeptide, demonstrating that it can survive rather hostile ionizing environments, or to the formation of longer linear peptide chains, thus playing a key role in the early stages of the chemical evolution of the life.

The study of other cyclic dipeptides like cyclo(glycine-alanine) and cyclo(glycine-glycine) provides the opportunity to probe the generality of the proposed mechanisms. The results show complex fragmentations far from trivial or generalisable from one species to another.



The 'smart decomposition' of cyclo(alanine—alanine) dipeptide under VUV irradiation

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Theoretical studies of reactive scattering processes involving the H_2 reaction complex

Scattering processes such as mutual neutralization, double charge transfer, associative ionization, dissociative recombination and resonant ion-pair formation involve highly excited states and non-adiabatic effects are therefore crucial. I will present a model that makes it possible to systematically study all of the above processes *ab initio* and fully quantum mechanically using accurate potentials and couplings. More specifically, the model makes it possible to include an accurate description of bound electronic states as well as Rydberg states and electronic resonant states. Furthermore, it includes non-adiabatic couplings between bound electronic states, both at small and large internuclear distances, as well as couplings to resonant states and to the ionization continuum. Results will be presented on an application of the model to the H_2 system. The model is not limited to the H_2 system and may serve as a basis for which more complicated systems can be studied.

Studies of molecular properties using the Self-Heterodyne effect using a chirped Quantum Cascade Laser

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The external-cavity quantum-cascade laser (EC-QCL) has become an important radiation source in the mid-IR (3μ to 20 μm) portion of the electromagnetic spectrum. QCLs have been successfully implemented into a variety of spectral studies for different applications, such as atmospheric and environmental monitoring [1], detection of trace explosives [2], and human breath analysis [3]. QCLs generate tunable laser light by means of a periodic series of nanometer sized layers which leads to one-dimensional multiple quantum well confinement. In the pulsed mode, a high electron current is injected into the active medium and, therefore, inducing joule heating. This in turn modifies the refraction index of the QCL medium leading to a fast wavelength tunning induced by the current pulse thus allowing "chirped" operation of the laser. In our work, we use an Uber Tuner, pulsed, broadly tunable EC-QCL from Daylight Solutions with a emission range in the mid-IR tunable from 6.9 to 8.7 μm. The pulse width can be selected in a range from 40 ns down to 500 ns with a variable repetition frequency of 100Hz down to 100 KHz. The QCL delivers a maximum pulse power up to 350mW close to the middle of the tunning range. A Herriot cavity is coupled to the laser and is placed in between the QCL and the MCT. In order to capture the time dependency of the QCL laser for a given frequency we used a SRS 2000 boxcar gated integrator with picosecond resolution. With this arrangement, we have achieved measuring Self Heterodyne Excitation of the H_2O molecule. This phenomenon occurs when a pulse with a fast frequency sweep is made to interact with a resonant system in the gas phase. A simple picture of the self heterodyne process can be obtained if one considers that the original pulse, travelling through the cavity, adds up linearly with the delayed pulse as produced by the molecular sample leading to interference patterns in the total radiation detected at the end of the cavity, if filled up with the sample gas[4]. A more complete analysis of this process requieres the analysis of a closed two level system interacting with a swept field, in terms of the optical Bloch equations. In the present work we report pressure dependent self heterodyne signals as produced in the Herriot cell by the resonant absorption of water vapor in the cavity corresponding to a rotation-rotation transition J-J´ in the vibrational ground state of the H2O. Beyond this specific use of Quantum Cascade Lasers, I will also discuss potential applications of Quantum Cascade Lasers in state selective autodetachment of negative ions, studies of the competition of detachment-dissociation channels in negative molecular ions and the potential use of intracavity Mid Infrarred Spectroscopy as a potential diagnostic tool in ion storage rings.

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Acknowledgements. The authors acknowledge the financial support of the PAPIIT-DGAPA project -IN116920

A gas-filled cryogenic Radio-Frequency Quadrupole (RFQ) ion beam cooler for laser photodetachment at GUNILLA

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Laser photodetachment threshold spectroscopy (LPT) [1] has been commonly employed in studies of negative ions at Gothenburg University Negative Ion Laser LAboratory (GUNILLA), where photodetachment cross sections are measured by recording the number of neutral atoms produced as a function of photon energy in a laser-ion merged-beams format. In recent years, ion beam bunchers based on a gas-filled linear Radio-Frequency Quadrupole (RFQ) traps have become a standard tool to improve beam quality, i.e. good emittance/low energy spread [2]. For continuous-beam experiments as performed at GUNILLA, a single-pass cooling scheme is more advantageous, which has been successfully developed and applied for isobar suppression in Accelerator Mass Spectrometry (AMS) at the world leading facility VERA (Vienna Environmental Research Accelerator) [3]. Another advantage of ion cooling is an increased photon-ion interaction time, which may significantly enhance the photodetachment efficiency. This will also greatly reduce the experimental uncertainties. In the goal of a breakthrough in photon-ion experiments, implementation of a cryogenic RFQ ion beam cooler using helium buffer gas is proposed.

This cooler will first reduce the kinetic energy of negative ions down to the few eV range by means of electrostatic deceleration. Negative ions then undergo multiple collisions with high purity buffer gas inside the quadrupole, eventually equilibrate to an energy that corresponds to the temperature of the buffer gas. The RF field in the quadrupole will create a transversal average trapping potential that forces the ions close to the central axis of the quadrupole. Additional four tapered electrodes between the RF rods provide a small gradient field to guide the ions downstream through the cooler.

The whole system will be operating at a cryogenic temperature (10 K or even less) in order to achieve a very low velocity spread of the ions and a smaller beam diameter. The helium pressure inside the RFQ cooler will be of the order of 0.1 mbar. To limit losses via collisional detachment, high-vacuum conditions are preserved immediately before and after the RFQ cooling section. Thus, a multi-stage differential pumping system with several skimmer diaphragms and high gas throughput will be implemented. This will also serve as a thermal shielding for the central interaction region.

The RF setup will have two resonant circuits coiled in opposite direction driven via a ferrite transformer by a 200 W RF power amplifier and a function generator. A binary set of 9 purpose-built switchable inductors ensures resonance conditions for different frequency tunings. It may achieve zero-to-peak voltages of around 400 V in the frequency range of 1.2−6 MHz, thus, allow optimal cooling of ions with masses between 14 and 350 u. For ions outside this mass range, the RF-voltage has to be adjusted accordingly.

Detail design of the cooler will be presented in this work. Furthermore, the possibility of utilizing such a cooler in the injection beamlines of DESIRREE will be discussed.

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Radioactive negative ions at ISOLDE

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Negative ions are unique quantum systems to probe electron correlation effects: since the Coulomb potential of the nucleus is almost entirely screened, the binding of the additional electron is primarily due to many body interactions between electrons. Consequently, negative ions are sensitive probes for electron correlation theories that go beyond the independent particle approximation, which is of crucial importance for the understanding of atomic and molecular structure. However, due to the weak binding potential, the energy gained by attaching an electron to a neutral atom, referred to as electron affinity (EA), is typically only of the order of one eV. For the same reason, negative ions typically lack bound excited states with opposite parity, with the noticeable exceptions being lanthanum, cerium, osmium and thorium [1-4]. Consequently, the EA is the only parameter which can be probed with high precision, typically via laser photodetachment threshold spectroscopy. Furthermore, the determination of isotope shift in the EA is an excellent benchmark to probe the accuracy of theoretical calculations of the specific mass shift, which is required when e.g. extracting nuclear charge radii from spectroscopic data.

In order to study radiogenic elements, in particular those with short half-lifes, an on-line facility such as CERN-ISOLDE needs to be utilized, where elements with half-lifes of >30ms can be produced and delivered to experimental setups. A program to study EAs and isotope shifts utilizing radiogenic negative ions at ISOLDE was established by the GANDALPH collaboration a few years ago, reaching a major milestone with the determination of the electron affinity of astatine utilizing the GANDALPH detector in 2018 [5]. Following the long shutdown of the CERN accelerator complex, experiments determining the isotope shift in the EA of chlorine as well as the determination of the EA of polonium are being prepared and are expected to take place in 2023.

Furthermore, efforts to convert positive ions delivered by ISOLDE to negative ions using the charge-exchange process were initiated, broadening the range and accessibility of negative ion experiments at ISOLDE. Initial yields of the production of uranium from charge exchange reactions by injecting a 40keV ion beam into a sodium filled charge exchange cell were performed in 2022.

Here, we will present the status and developments of the negative ion program at ISOLDE including the measurements of the EA of ^{128}I and ^{211}At, charge exchange production yields as well as future activities including the isotope shift in the EA of chlorine, the EAs of Po, Fr and the actinide elements.

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The optical atom: A bridge between atomic physics and optics

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(Dated: June 28, 2022)

The models used to describe the properties and behavior of atoms are based on several parameters fixed by nature, such as the elementary charge or Plank's constant. Ideally, we would like to be able vary these parameters and observe how the properties of the atoms changes. In this contribution we show how the optical properties of an evaporating droplet can be used to mimic an atom with a varying radius. We measured the Directional Mie Scattering of the evaporating droplets, thus cleaning the spectrum and selecting the TM modes. We observe a series of Fano resonances arranged in a comb structure, as the droplet evaporates. The explanation uses an analogy between electromagnetic light scattering and the Schrödinger equation. The effective potential in the water droplet can be described as an attractive square well potential where the step in the potential occurs at the radial boundary of the droplet. The angular momentum of the electron in an atom is here replaced by the angular momentum of light rotating inside the droplet. In the experiment we sweep in droplet size, equivalent to atom size, as the droplet evaporates. Each comb structure is related to different excited states and the resonances inside a comb are given by consecutive angular momentum numbers ℓ. The Fano nature is a consequence of tunneling through the potential barrier. From this point of view, we generate an experimental simulator that represents an atom with a knob to control its size.

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The search for non-IPR isomers of C_{60}^{-} in DESIREE

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After the accidental discovery of fullerenes [1] and its unequivocally identification in the interstellar media (ISM) [2], extensive studies were made to elucidate their specific molecular geometries. These were found to be closed cages with an even number of carbon atoms arranged as 12 pentagons and zero, two or more hexagons. C_{60} is the smallest member of the fullerene family that obeys the so-called isolated pentagon rule (IPR) [3], where each pentagon is surrounded by 5 hexagons for the most stable isomer. This geometry gives the molecule its exceptional stability and chemical inertness, making it the most abundant of the fullerenes. Every other C_{60} isomer has at least two pairs of adjacent pentagons. For the first non-IPR isomer of the C_{60}- fullerene, the two adjacent pentagon configuration leads to an enhanced reactivity and a significantly higher electron affinity [4]. The latter suggests that this isomer could have a significant survival probability in astrophysical environments.

A previous study performed in the cryogenic electrostatic ion storage ring facility DESIREE also supports this idea. In this storage ring, the spontaneous decay of hot molecules can be followed on timescales up to hundreds of seconds owing to its excellent vacuum conditions (10^4 cm^{-3} of residual gas density [5]). The observed decay in the case of C_{60}- at timescales exceeding milliseconds could not be explained by the presence of the IPR isomer alone (Figure 1), which suggests that a small population of the first non-IPR isomer of C_{60}-, which do not cool efficiently, is being produced. The presence of this isomer could be measured in DESIREE by laser probing the depletion of neutral yield as a function of storage time.

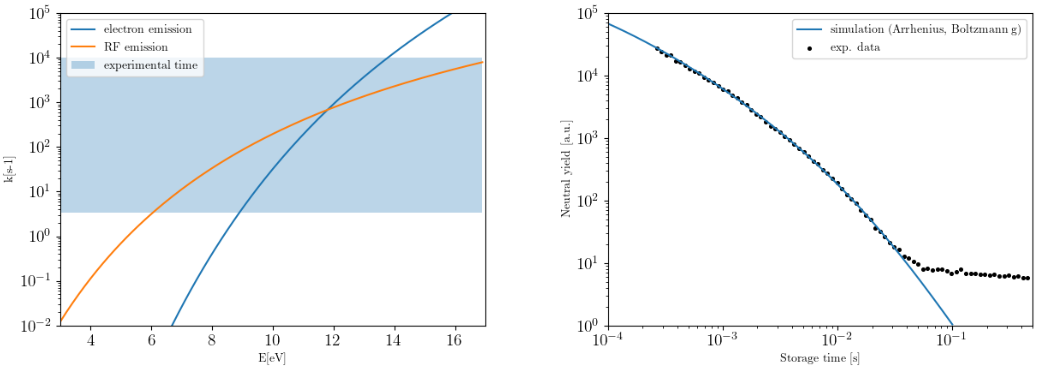

Figure1.- The left panel shows the significant rate constants as a function of internal excitation energy for the IPR C_{60}-. The blue curve describes the electron emission from vibrationally hot anions while the orange curve shows the recurrent fluorescence emission rate. The blue shaded region indicates the time window of the measurement (100μs – 500 ms). The right panel shows the measured neutral yield as a function of storage time (black data points) together with the corresponding results from our simulation.

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Mutual Neutralization of H_3O^+ + OH^- experiments at DESIREE

One of the basic mutual neutralization (MN) reactions between molecular ions is the MN of hydronium and hydroxide, which is the inverse autoionization of water:

$$2H_2O \rightleftharpoons H_3O^+ + OH^-$$

The balance between these two proton-transfer reactions defines the pH of pure water. However, mutual neutralization of the two ions, isolated from their environment can be considered to most likely proceed via electron transfer, forming an unstable H_3O^* and an OH radical. Recent development of imaging the neutral products of cold MN reactions at the DESIREE facility made it possible to detect the products of such isolated MN reactions. Indeed, we attribute the dominant mechanism to electron transfer, followed by dissociation of the unstable H_3O^* product either into H_2O + H or to OH + H_2, observed along with a neutral OH radical as an overall three body breakup of the neutral complex. Nevertheless, we observe a significant contribution of two body H_2O + H_2O breakup, which can only be attributed to a proton-transfer mechanism. In the meeting we will present and discuss the kinetic energy release distributions of the different product channels that allows to determine the degree of internal energy deposited in the internal degrees of freedom of the molecular products. Furthermore, the long storage times allow to observe the change in the relative e-transfer and proton-transfer yields as a function of storage time and cooling of the ions in the 13K ambient temperature at DESIREE.

Mutual Neutralization in sub-eV $C_{60}^{+} + C_{60}^{-}$ collisions

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The detection of fullerenes in neutral and ionic forms in several extra-terrestrial environments [1][2][3] has been subsequently followed by extensive research, inquiring about the factors that constitute the molecule's formation and survival in these environments [4]. To advance the understanding of charge transfer processes involving fullerene molecules and their importance in e.g. space, we have performed mutual neutralization studies using the cryogenic storage ring facility, DESIREE [5]. With DESIREE, we are able to study sub-eV collisions between C_{60}^{+} and C_{60}^{-} ion beams, which marks the first-ever reported studies in this regime for molecular collisions. We detect neutral products formed in the process, that have an energy distribution with identifiable electronic excited states. The process is then modelled with the multi-state Landau-Zener method [6], using relevant interaction potentials [7][8] and coupling elements [9]. The excited states of C_{60} used in the model were computed using Time Dependent-Density Functional Theory at the PBE0/6-311++G(d,p) level of theory. The Landau Zener model predicts the total charge transfer cross section to be 5.52×10^{-11} cm^{2}, for a center-of-mass collision energy of 1 meV. This is a factor of 4-5 larger than for $H^{+} + H^{-}$ collisions at the same collision energy [10]. The branching ratios subsequently obtained from the theoretical calculations follow the experimental trend observed for the same.

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Highly-sensitive photodetachment spectroscopy in an MR-ToF device

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The electron affinity (EA) reflects the released energy when an electron is attached to a neutral atom. An experimental determination of this quantity can serve as an important benchmark for atomic models describing electron-correlation effects [1]. A comprehensive understanding of these effects is also necessary for accurate calculations of the specific mass shift, which is required to extract nuclear charge radii from measurable total isotope shifts. By measuring small differences in the EA between different isotopes of the same chemical element, the isotope shifts of the EA, atomic models can be further constrained. However, isotope shifts in the EA have been experimentally determined only for very few stable nuclides so far, and only with modest precision. As an example, the isotope shift between the two stable chlorine (Cl) isotopes is more precisely predicted in theory [2] than experimentally measured [3].

Exploiting the low-energy version of the Multi Ion Reflection Apparatus for Collinear Laser Spectroscopy (MIRACLS) [4], we have initiated a high-precision measurement of the isotope shift in the electron affinity between stable Cl isotopes as well as the long-lived ^{36}Cl isotope. This can be achieved by photodetachment threshold spectroscopy of negative Cl ions. By trapping ion bunches between the two electrostatic mirrors of MIRACLS' multi-reflection time-of-flight (MR-ToF) device, the same ion bunch can be probed by the spectroscopy laser repeatedly. As a result, the photodetachment efficiency can be significantly increased in comparison to single-pass experiments. Thus, instead of conventionally used pulsed high-power lasers with a large linewidth, narrow-bandwidth continuous-wave (CW) lasers can be employed. Consequently, the measurement precision will be improved.

By confining the Cl^- ions for a few 10,000s of revolutions in the MR-ToF device, neutralised atoms produced by photodetachment have been experimentally detected. For wavelengths only 3 nm above threshold, a CW laser power as low as 0.8 mW has been demonstrated to be sufficient to observe the process of photodetachment. The first experimental data indicates that the new method is 3 to 4 orders of magnitude more sensitive than conventional single-pass photodetachment experiments, see e.g. [1]. Thus, our novel measurement scheme paves the way for precision measurements of isotope shifts in the electron affinity between stable and ultimately radioactive Cl isotopes.
As such, the MR-ToF technique can become complementary to recently reported high-precision photodetachment measurements at storage rings such as DESIREE [5] and, furthermore, extend the range of isotopes beyond the stable ones.

Due to its small floor space of just 2m x 1m, an MR-ToF apparatus can be easily installed at existing radioactive ion beam facilities. Combined with higher power lasers, the MR-ToF technique will hence allow measuring EAs and isotope shifts in the EA for various radioactive negative ions for the very first time.

The novel technique will be introduced and first experimental results will be presented.

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Efficient stabilization of cyanonaphthalene by fast radiative cooling

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After decades of speculation and searching, astronomers have recently identified specific Polycyclic Aromatic Hydrocarbons (PAHs) in space. Remarkably, the observed abundance of cyanonaphthalene (CNN, $C_{10}H_7CN$) in the Taurus Molecular Cloud (TMC-1) is six orders of magnitude higher than can be explained by astrochemical modeling. Here, we report absolute unimolecular dissociation and radiative cooling rate coefficients of the 1-CNN isomer in its cationic form. These results are based on measurements of the time-dependent neutral product emission rate and Kinetic Energy Release distributions produced from an ensemble of internally excited 1-CNN$^+$ studied in an environment similar to that in interstellar clouds. We find that Recurrent Fluorescence – emission of optical photons from thermally populated electronic excited states – efficiently stabilizes 1-CNN$^+$, owing to the large enhancement of the electronic transition probability by vibronic coupling. Our results help explain the anomalous abundance of CNN in TMC-1 and indicate that small PAHs may be stable in molecular clouds and in the interstellar medium in general, challenging current understanding in astrochemistry.

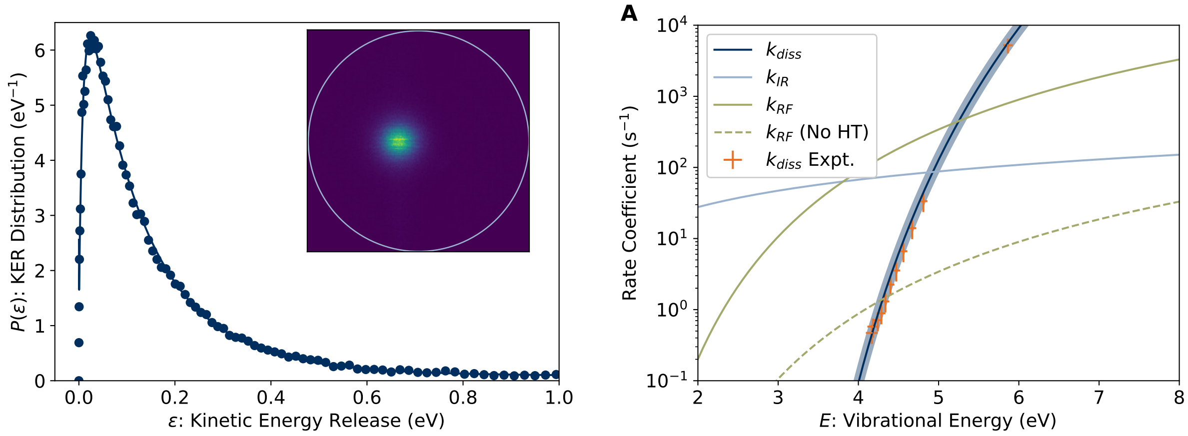

Figure 1: Left: Kinetic Energy Release distribution for 1-CNN$^+$ dissociating 120 μs after formation, determined from the inset detector image. Right: Unimolecular dissociation and radiative cooling rate coefficients for 1-CNN$^+$.

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Radiative lifetimes in atomic negative ions

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Abstract

The first new scientific output from the Double ElectroStatic Ion Ring ExpEriment (DESIREE) was a measurement of the lifetime of the metastable $^2P^o_{1/2}$ level in $^{32}S^-$ [1]. Since then, the lifetimes of excited states in several atomic negative ions have been measured. The method has also been applied at the Cryogenic Storage Ring (CSR) in Heidelberg where Si^- was investigated [2].

Transitions between bound states in atomic negative ions are in most cases forbidden. This leads to very long lifetimes of the excited states, which only can be investiated if the ions can be stored for long times. The lifetimes are measured using the laser photodetachment spectroscopy technique where an OPO laser is used to photodetach only the excited state(s) in the ion. The ions are produced in a sputtering process which yields very high populations in excited states. The lifetime can be probed measuring the population of the excited state as a function of the time after injection into the ring. These experimental results can be used to benchmark theoretical methods that go beyond the independent particle model, since such models break down in the case of negative ions.

Some of the ions that have been studied are Ir^-, Bi^-, Rh^-, Sb^- and W^-. One of the ions that has been studied recently is $^{75}As^-$, where two excited states were observed. The preliminary results indicate a $^3P_1$ state with a lifetime of 43.1(11) s shown in figure 1.

I will in this work present the experimental method and the give a brief overview of the work done so far.

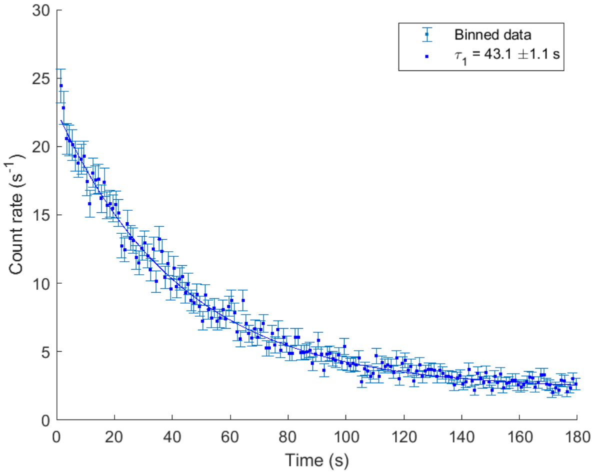

Figure 1: Preliminary results for the lifetime measurements of one of the $^3P_1$ in $^{75}As^-$.

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New Lifetime Limit for the Ground State Vinylidene Anion $\mathrm{H_2CC^-}$

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The molecular isomers acetylene (HCCH) and vinylidene $(\mathrm{H_2CC})$ form one of the simplest systems for studying isomeric reactions involving hydrogen.

In anionic form, the state with lowest total energy has the vinylidene structure, $\mathrm{H_2CC^-}$, with an electron affinity of about 0.5 eV. It lies 1.5 eV above the lowest neutral level of the acetylene structure, opening up the possibility that isomerization linked with electron emission could limit the lifetime of $\mathrm{H_2CC^-}$.

Indeed, an experiment at a room-temperature storage ring reported a finite intrinsic lifetime of about 110 s [1], by extrapolating from collision-limited (about 10 s) to collision-free beam lifetimes of $\mathrm{H_2CC^-}$, using a stable reference ion.

To access longer ion beam lifetimes, and thereby enable a better estimate of the ground state $\mathrm{H_2CC^-}$ lifetime, we employed the Heidelberg electrostatic Cryogenic Storage Ring (CSR) [2]. The CSR provides a cryogenic environment with strongly suppressed blackbody radiation and extremely low residual gas density, allowing to store and observe fast ion beams over time scales on the order of an hour.

We used photodetachment to monitor the decays of simultaneously stored $\mathrm{H_2CC^-}$ and $\mathrm{CN^-}$ ion beams. Here, $\mathrm{CN^-}$ served as a stable reference ion with nearly identical mass-to-charge ratio. Comparing the two decays at storage times of up to 3000 s allowed us to discriminate against most of the storage-ring induced loss processes. Furthermore, by using the novel isochronous mass spectrometry method [3], we were able to prove that no ion beam contaminations could significantly affect the observed lifetimes.

We present preliminary results which suggest that the intrinsic lifetime of the ground-state of $\mathrm{H_2CC^-}$ is several thousand seconds, i.e., at least an order of magnitude longer than assumed previously.

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Experimental confirmation of the isotope effect on the branching in mutual neutralization

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Synopsis We studied the mutual neutralization of $^{1,2}H^-$ with $^7Li^+$ and $^{16}O^+$ at effective collision temperatures of up to 2000 K, which corresponds to few hundred meV, and obtained branching fractions that show the theoretically predicted yet experimentally unobserved isotope effect.

Hydrogen is the most abundant element in the universe and, as such, one of the most important collision partner in many astrophysical plasma environments, such as e.g. stellar atmospheres [1]. The mutual neutralization with the hydrogen anion can be significant in the accurate determination of element abundances by non-local thermodynamical equilibrium modelling of stellar spectra. This has been demonstrated for the case of Li and Na lines [2, 3]. The low mass of hydrogen poses a technical challenge and hydrogen is often replaced with its heavier isotope, deuterium, in experiments. Hence, it is crucial to understand the isotope effect when experimental data on deuterium shall benchmark theoretical results on hydrogen, particularly at low collision energies of a few hundred meV, which correspond to the typical temperatures in stellar photospheres. As the overall isotope effect is a combination of multiple, opposing isotope effects, its sign and magnitude on absolute and partial cross sections is not easily predicted. In the case of neutralization of $^{1,2}H^-$ with Li^+, all theoretical studies at the energies of interest predict the presence of an isotope effect on both cross section and branching fractions [4, 5, 6]. Our recent experimental results of this collision system confirm the theoretically predicted presence and magnitude of the isotope effect on the branching fraction. Additionally, we pushed our instrumental limits on the reactants' mass ratio and were able to study the mutual neutralization of $^{1,2}H^-$ and O^+. Our results show a strong branching into $O(2s^2 2p^3 3p\ ^3P)$ with a kinetic energy release of 1.88 eV, which stands in contrast to previous experimental results on $^2H^-$ and O^+ [7].

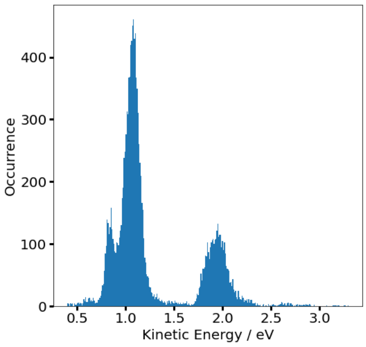

Figure 1. Distribution of the kinetic energy of products from the mutual neutralization between $^1H^-$ and O^+.

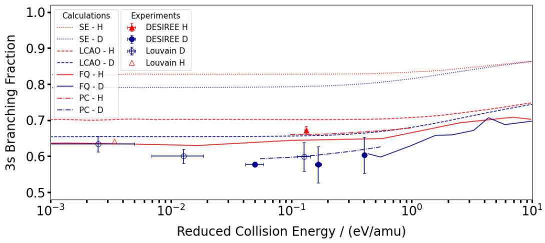

Figure 2. Experimental and theoretical branching fractions into $Li(1s^2 3s)$ for mutual neutralization of $^{1,2}H^-$ with Li^+ at different collision energies.

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The Largest Fullerene

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The ground state allotrope of bulk carbon, at temperatures below a few thousand Kelvin and pressures below a few GPa, is graphite [1]. At sizes of tens to hundreds of atoms, fullerenes instead stand out as a particularly stable allotrope [2]. This leads to the question of at what size graphitic materials become thermodynamically favorable over closed-cage fullerenes? Or, in other words, what is the largest fullerene? Here, we have used classical molecular dynamics to investigate this by modeling single- and multi-layered structures of graphene and fullerenes. Of the systems we have studied, single-caged fullerenes are the most stable structures for sizes up to about 10^4 atoms. So-called carbon onions [3], multi-layered fullerenes then become the most stable allotrope at sizes above this and only at sizes above 10^{11} atoms do graphite-like structures become the energetically preferred species. I will present a detailed overview of our calculations and findings.

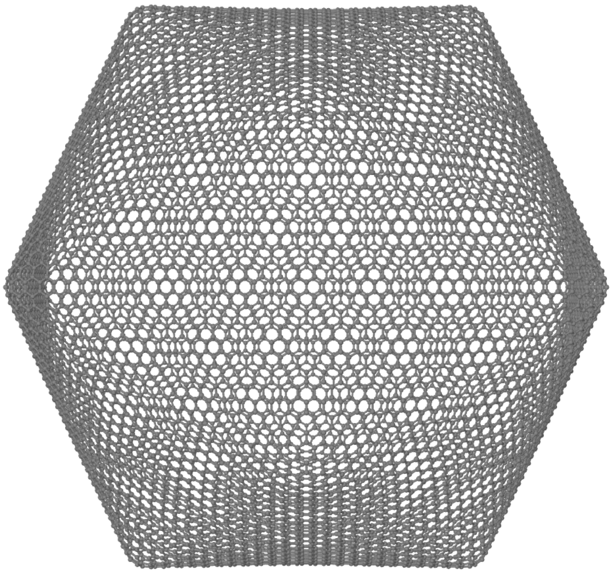

FIG. 1: The optimized structure of the C_{10140} fullerene.

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High-precision electron affinity of oxygen

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Negative ions are important in many areas of science and technology, e.g., in interstellar chemistry, for accelerator-based radionuclide dating, and in antimatter research. They are unique quantum systems where electron-correlation effects govern their properties. Atomic anions are loosely bound systems, which with very few exceptions lack optically allowed transitions. This limits prospects for high-resolution spectroscopy, and related negative-ion detection methods. Here, we present a method to measure negative ion binding energies with an order of magnitude higher precision than what has been possible before. By laser-manipulation of quantum-state populations, we are able to strongly reduce the background from photodetachment of excited states using a cryogenic electrostatic ion-beam storage ring where keV ion beams can circulate for up to hours. The method is applicable to negative ions in general and here we report an electron affinity of 1.461 112 972(87) eV for ^{16}O. This unprecedented high precision opens up for studies of isotope shifts at levels of detail that will challenge atomic and nuclear theory. The isotope shift in the electron affinity of $^{18-16}$O will also be presented.

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The Life, Death and Knockout of Interstellar Carbon

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Polycyclic aromatic hydrocarbons (PAHs) and fullerenes, two families of carbon-based molecules, both present in the interstellar medium, have since the 1980's been of particular interest for astrophysicists. The existence of PAHs in space was proposed at that time [1][2], but the first unique species was not discovered until 2021 [3]. Fullerenes – the most well-known fullerene being the large, football-shaped C_{60} molecule – were first observed in experiments [4] and later also in space [5]. Their interstellar presence aside, these two types of molecules have in common that they provide unique insights into knockout processes that lead to them losing individual carbon atoms in energetic collisions.

To further understand the creation and longevity of such "imperfect" molecules, we collided C_{60} and PAH coronene respectively with He atoms and studied the remaining products. This was done experimentally at the DESIREE facility, colliding 30 keV C_{60} molecules with stationary He atoms, as well as by simulation, using the LAMMPS molecular dynamics software. In both experiment and simulation, in the C_{60} as well as the coronene case, we saw that it is possible for C_{59} and PAH knockout fragments to be created - this was known from previous experiments [6], but they had so far only been shown to survive on microsecond timescales. While our simulations show the creation rate and early stages of the C_{59} and PAH fragments, our experiments show that the fragment population has cooled down significantly on a 10 ms timescale and remain stable thereafter [7][8]. Thus, we conclude that it is possible for such molecules to survive indefinitely in the gas phase.

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Cooling dynamics of nitrogen-containing PAH cations (PANHs)

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Polycyclic aromatic hydrocarbons (PAHs) are a class of organic molecules based on two or more fused aromatic rings, with hexagonal rings as typical dominant constituents. They are found and formed in a wide variety of environments. In the case of extraterrestrial environments, PAHs are believed to be an important component in interstellar dust and gas and as such are responsible for infrared emission features that dominate the spectra of many galactic and extragalactic sources [1]. Numerous experimental and theoretical studies on PAHs have been carried out in the past decades, but how they may survive in space is still not fully understood [2].

PAHs that contain one or more nitrogen atoms are so-called polycyclic aromatic nitrogen heterocycles (PANHs). The existence of PANHs in space has been inferred by their detection in meteorites [3], and they may be present in the haze surrounding Saturn's largest moon Titan [4]. Their presence in astrophysical environments is further supported by the recent discoveries of two interstellar nitrile group functionalized PAH molecules (1- and 2- cyanonaphthalene) [5] and one of the simplest nitrogen-bearing aromatic molecules (benzonitrile: c-C_6H_5CN) [6] in the TMC-1 molecular cloud. One key issue is the cooling dynamics of such molecules, which affects their abundances in the ISM [7].

The ultra-high vacuum and low-temperature environment provided by the DESIREE ion-beam storage ring facility [8] is ideal for following ultraslow molecular relaxation processes of astrophysical interest on timescales exceeding seconds, where fragmentation, recurrent fluorescence, and IR emission compete. In this talk, I will present such studies of PAH molecules with a focus on the spontaneous and laser-induced decays of acridine ($C_{13}H_9N$) and phenazine ($C_{12}H_8N_2$) cations. Both molecules are members of the PANH family and consist of three planar aromatic rings but differ in that a C–H groups in the central anthracene ring have been replaced by one N to form acridine or two N to form phenazine. Fig.1 shows examples of the spontaneous decays of hot acridine and phenazine cations as monitored through the yields of neutrals due to unimolecular dissociation products as functions of ion beam storage time. These curves provide information on the characteristic radiative cooling times for which the decays are quenched, which together with results using laser probing techniques allow following the cooling dynamics in unprecedented detail.

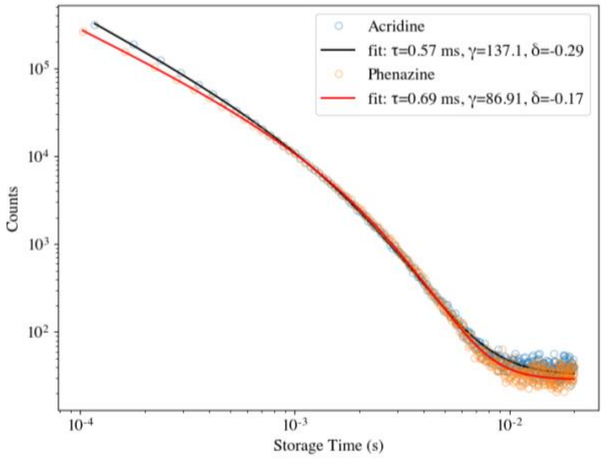

Figure 1: Spontaneous decay of acridine and phenazine cations as a function of storage time.

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A versatile ion source for cold ions using superfluid helium nanodroplets

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Superfluid helium nanodroplets (HNDs) can be used as a nano laboratory to study atoms, molecules and clusters at cryogenic temperatures [1]. Recently, it was shown that it is possible to highly charge these HNDs without breaking them [2]. Furthermore, it was shown that these charges efficiently ionize almost every atom or molecule, picked up by the HNDs, acting also as nucleation centers for the formation of small ionic clusters [3,4]. Based on these results we constructed a new experimental setup, which can be used as an intense ion source for both, positively and negatively charged ions.

HNDs are produced by supersonic expansion of precooled (below 10K) and pressurized (about 20 bar) He 6.0 in ultra-high vacuum. These droplets are then ionized by electron bombardment, in a crossed beam electron impact ion source forming highly charged droplets. In the pick-up region, these HNDs can pick up almost any atom or molecule by individual collisions. To liberate the low-mass ions from the HNDs a movable and isolated target is place in the path of the droplets. A splashing like behaviour during the collision with the surface allows the extraction of the majority of the ions embedded in the HNDs. These ions are then guided to a conventional high-resolution time of flight mass spectrometer (QTOF-premier, Waters) with which the resulting cluster size distributions can be analysed. In addition, a quadrupole mass filter allows us to select a specific ion and further investigate it specific properties by additional collisions with atoms or electrons or perform helium tagged action spectroscopy measurements. Furthermore, a high ion yield would allow us also to store these in the double electrostatic ion ring experiment (DESIREE) and perform various experiments.

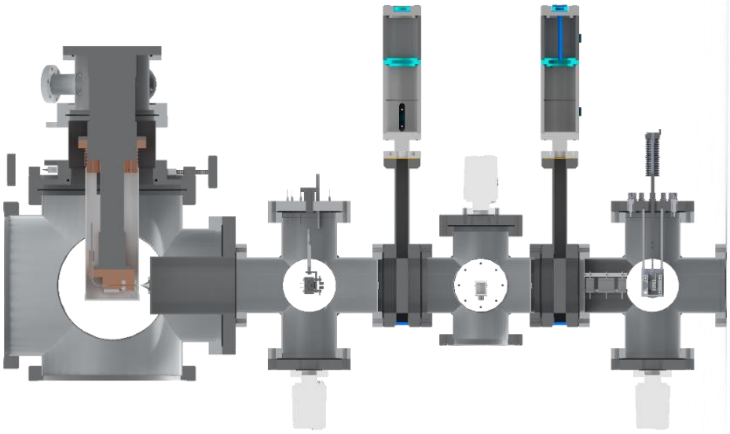

*Figure 1 Rendering of the newly developed experimental setup consisting of several differentially pumped sections. Starting from the left with the helium cluster source, which if followed by an electron impact ion source to highly charge the HNDs. The pick-up chamber is used to pick up almost any atom or molecule. On the right a special designed polished stainless-steel target is placed in the path of the HNDs and is used to extract low mass ions from the HNDs and guide them towards the conventional time of flight mass spectrometer (not shown here)***.***

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Charge transfer in Sodium Iodide collisions

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Sodium Iodide (NaI) have over the years served as a prototype system in the studies of avoided crossing dynamics. Here, the charge transfer collision reactions Na+I $\leftrightarrows$ Na^++I^- (mutual neutralization and ion-pair formation) are studied from an *ab initio* approach and the total- and differential cross sections are calculated for the reactions. This involves electronic structure calculations on NaI to obtain adiabatic potential energy curves, nonadiabatic coupling and spin-orbit coupling followed by nuclear dynamics, treated fully quantum mechanically in a strict diabatic representation. A semi-empirical spin-orbit coupling model is also investigated and the effects of rotational coupling is estimated. The calculated total- and differential cross sections are compared with measured ion-pair formation cross sections and analysed via the semi-classical Landau-Zener model.

Laser probing of metastable Ba$^+$ for lifetime measurements

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Metastable levels are responsible for parity forbidden lines occuring in many low-density astrophysical plasmas, found in e.g gaseous nebulae, planetary nebulae, protostars, stellar chromospheres. Line ratios from forbidden lines are the most reliable tools for diagnostics of temperatures and density of these regions. Measurements of metastable lifetimes is direct importance for the use of forbidden lines. Such a lifetimes can be about 100sek long and therefore the low temperature and low pressure experimental chambers are demanded in order to avoid the depopulation of metastable levels due to collisions and thermal radiation. The facility Double ElectroStatic Ion-Ring ExpEriment, DESIREE [1, 2] at Stockholm University demonstrated capacity for such a measurements.

The laser probing technique (LPT) was derived by Mannervik and his group at the CRYING storage ring, and successfully applied to a number of ions of varying complexity [3] For several complex ions, the measured lifetimes were combined with astrophysical line ratios to derive experimental transition rates [4].

We propose to further develop the laser probing technique to measure lifetimes for stored positive ions using DESIREE. One of the most favorable ions to develop the technique of laser probing of a stored ion beam is Ba$^+$. The atomic structure is simple with few levels and the metastable energy levels are located at low excitation energies. This allows for a high population and increased fluorescence signal, making Ba$^+$ an ideal target ion.Recently Lifetime of Ba$^+$ metastable state 5d^2 D$_{5/2}$ was measured in Paul trap [5] and this can be used as reference

In our poster we will report a results of our first effort in the development of a LPT for DESIREE, and will discuss the next proposed experiment, what we are going to start on DESIREE in August 29, just after this Symposium

Acknowledgements
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Ion source based on combined RF frequency powered inductively coupled plasma and hollow cathode discharge

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Radiofrequency powered inductively coupled plasma discharge *(RF-ICP)* is a well-known source of very intensive atomic resonance spectra where resonance lines are up to 10 times more intensive /1/.
There are still problems in production spectra sources for atomic analytical spectroscopy, but ICP is widely applicable in sophisticated experimental research /2,3,4/. Hollow cathode (HC) discharge is broadly used commercially in atomic analytical instrumentation to get atomic resonance spectra line for atoms of many elements of Mendeleev periodic system including hardly volatile elements, e.g. boron. HC lamps alongside atomic emmit sufficiently intensive resonance lines of ionized atoms, which rarely is the case for RF ICP plasma.

Based on our long-term expertise in manufacturing of various light sources /5,6/ we designed the device where both RF-ICP and HC plasma forms a hybrid system. The coil of the RF-ICP source is positioned just next to the cathode of the modified commercial hollow cathode lamp. As a result, we obtained amplification of intensities of emitted HC spectra by 3-5 times for atomic and ionic resonance lines of the selected element. We applied the hybrid system for Cd and Zn because their atomic and ionic resonance lines are closely positioned in the UV spectra region (210-240nm), and it is very suitable to measure and optimize the ratio of the number of atoms to the number of ions in the plasma. Presence of intensive ionic spectra lines evidences the abundance of ions for selected elements in the plasma produced by the hybrid system.

We adapted our hybrid system to serve as an ion source and attached it to our ion beam setup GRIBA (Gothenburg Riga Ion Beam Apparatus) /7/. Currently, the investigations of the positive ion flow extracted from our hybrid ion source are on the agenda in dependence on gas flow, the power of the HC lamp and RF-ICP source, and the geometry of the position of RF-ICP coil and HC.

Our main interest is to apply such a hybrid system for the generation of the boron ion beam, which could be implemented into an innovative implantation device. This work is inspired by Baltic Scientific Instruments /8/, worldwide known as a producer and supplier of ionizing radiation detectors based on high purity Germanium crystals.

Acknowledgement. This work is supported by ERDF project No. 1.1.1.1/19/A/144 “Technologic rese-arch for elaborating the next generation boron ion implantation apparatus with TRL level near to 4"

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A community platform for just atomic computations

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Electronic structure calculations of atoms and ions have a long tradition in physics with applications in basic research and spectroscopy. With the Jena Atomic Calculator (JAC), I here present a new implementation of a (relativistic) electronic structure code for the computation of atomic amplitudes, properties as well as a large number of excitation and decay processes for open-shell atoms and ions across the periodic table. JAC [1] is based on Julia, a new programming language for scientific computing, and provides an easy-to-use but powerful platform to extent atomic theory towards new applications.

A primary guiding philosophy in designing JAC was to develop a general and easy-to-use toolbox for the atomic physics community, including an interface that is equally accessible for working spectroscopists, theoreticians and code developers. In addition, I also wish to provide a modern code design, a reasonable detailed documentation of the code and features for integrated testing [2].

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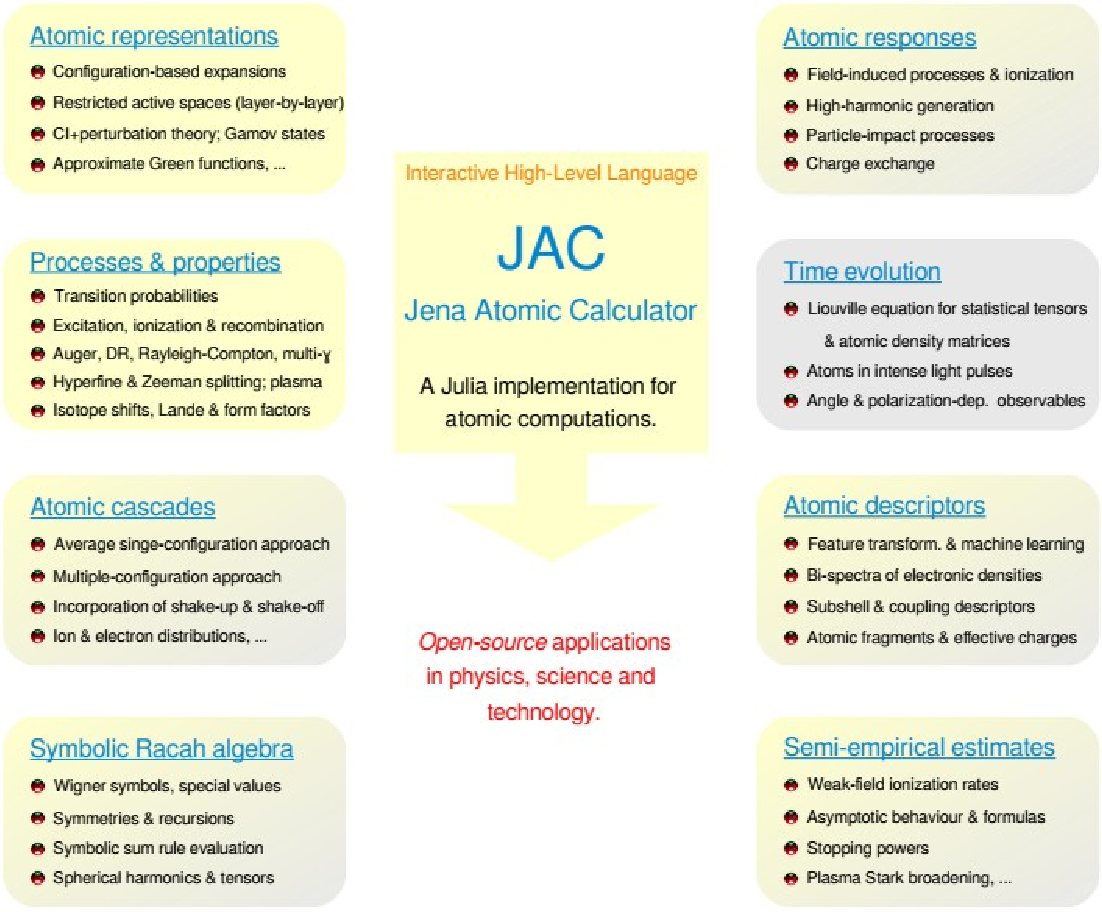

Dissociative recombination of internally cold CH^+ molecules

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Heavy ion storage rings provide an ideal environment for internal state preparation of atomic and molecular ions and subsequent collision experiments over a large range of collision energies using the merged-beams technique. We have studied the dissociative recombination (DR) of CH^+ in the electrostatic Cryogenic Storage Ring (CSR; [1]). In the cryogenic environment of CSR ($T < 20$ K), CH^+ ions radiatively decayed toward their lowest rovibrational states [2,3] and the internally relaxed ions were used for DR experiments. Merging an electron beam in the CSR electron cooler with the ion beam, low energy (meV) collisions could be probed. Measurements for different internal state populations enabled us to extract the rate coefficient for the ($v=0$, $J=0$) ground state.

DR reactions for $CH^+(v=0, J=0)$ at meV collision energies are an important part of the chemistry in diffuse interstellar clouds, out of which stars and planets can form [4]. The DR rate coefficient is needed to model this chemistry and interpret astronomical observations. Theoretical calculations are not yet reliably able to produce the required DR data due to the large number of intermediate states involved in the dynamics. Thus, laboratory studies of DR are needed to understand the CH^+ chemistry. With our experimental results at diffuse cloud conditions, we have significantly increased the reliability of the CH^+ diffuse cloud astrochemical models.

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On absolute dating with ^{14}C and ^{41}Ca

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^{14}C

It is well-known that ^{14}C ($t_{1/2}$ = 5,700 yr) requires a calibration curve to determine an absolute age from the measurement of the ^{14}C content in a material to be dated. Global calibration curves have been established for the entire ^{14}C dating range of about 50,000 years from the measurement of ^{14}C in absolutely dated archives [1-3]. However, sometimes 'plateaus' in the calibration curve hamper considerably the determination of a precise ^{14}C date, such as for the famous Minoan eruption of the Greek Island of Santorini some 3500 years ago [4].

Direct absolute ^{14}C dating would require the measurement of both ^{14}C and its stable beta-decay product ^{14}N*. The age of an object can thus be determined from a measured abundance ratio of ^{14}N*/^{14}C and the known half-life. Although this is considered to be impossible because the feeble radiogenic ^{14}N* signals are always overwhelmed by the omnipresence of nitrogen on Earth, an attempt at absolute ^{14}C dating has been discussed before [5].

^{41}Ca

Another radioisotope of considerable interest for archaeological dating is ^{41}Ca ($t_{1/2}$ = 100,000 yr). Bones contain a considerable amount of Ca, and the longer half-life would be ideal for dating the remains of hominides back to about 1 million years. This would make radiocalcium dating a particularly interesting tool [6]. Since a global calibration curve cannot be established for ^{41}Ca (it doesn't form a globally distributed gas like $^{14}CO_2$ from cosmogenic ^{14}C in the atmosphere), absolute dating through measurement of ^{41}Ca/^{41}K* ratios would be required. Again, there is the problem of the omnipresence of potassium on Earth.

In this presentation we will discuss the possibility for absolute dating of ^{14}C and ^{41}Ca by utilizing the kinematics of the beta decays leading to recoil energies of ^{14}N* and ^{41}Ca* of only $\leq$6.9 and $\leq$2.2 eV, respectively. A major question in the decay process is the probability of retention of the radiogenic isotopes within the original molecule, e.g. benzene ($^{12}C_5\mathbf{^{14}C}$) changing to $^{12}C_5\mathbf{^{14}N^*}$ or bone hydroxyapatite ($^{40}Ca_4\mathbf{^{41}Ca}$ $(PO_4)_3OH$) changing to $^{40}Ca_4\mathbf{^{41}K^*}$ $(PO_4)_3OH$.

^{14}C and ^{41}Ca can both be measured by accelerator mass spectrometry (AMS). But assuming that a retention of the decay products is likely, a sensitive method of detecting the minute amounts of stable radiogenic isotopes has to be found. We hope for stimulating discussions on these questions at the Symposium.

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