4.4 Superheavy elements at GSI and HI Mainz

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In 2023, there was no user beamtime served at GSI; activities at GSI therefore focused on the analysis and publication of data obtained previously, both at GSI as well as abroad, e.g., at ANU Canberra. Online chemistry studies were performed at NPI CAS Řež (CZ). Technical and method developments as well as offline work were performed at GSI and at HIM, for example for applications in laser spectroscopy, where a three-month campaign with the 20-h isotope ^{255}Fm obtained from a 40-d ^{255}Es generator system was employed to support two different studies: on the one hand this allowed complementing laser spectroscopy studies along a long isotopic Fm sequence by this neutron-rich isotope, and on the other hand it enabled fundamental studies in the life sciences. Technical developments to break current frontiers in future beamtimes to increase production rates, to advance to heavier, more exotic systems, to gain access to new observables and to provide higher-quality data were carried out.

Highlights in 2023

Synthesis / Nuclear Reactions

The heaviest superheavy elements (SHE) with atomic numbers Z=114-118 have been discovered in ^{48}Ca-induced fusion reactions with targets of Pu-Cf isotopes. Cross sections to form SHE in such fusion reactions decrease smoothly with increasing Z of the compound nucleus. The dynamical mechanisms underlying this trend are commonly attributed to an increase in the quasi-fission component acting prior to complete fusion. The quasi-fission outcomes from such reactions result in the emission of heavy fragments with masses around the doubly-magic lead, which have been studied at TASCA [A. Di Nitto et al., Phys. Lett. B 784, 199 (2018)] indicating the importance of the shell effect in the reaction dynamics. The origin of the effects of the ^{208}Pb shell closure was studied in collaborative work of the SHE Chemistry departments at GSI and HIM and the nuclear reactions group at the Australian National University (ANU), Canberra, Australia, at the Heavy Ion Accelerator Facility of ANU [1]. The ^{50}Ti projectile, most promising for the synthesis of elements beyond Og [J. Khuyagbaatar et al., Phys. Rev. C 102, 064602 (2020); H. M. Albers et al., Phys. Lett. B 808, 135626 (2020)] was used in studies in combinations with various actinide targets from ^{238}U to ^{249}Cf. In this work, the impact of the ^{208}Pb closed shells on the mass-distribution yield of quasi-fission products has been investigated. The results reveal that the sequential fission process could be responsible for the origin of the heavy fragments around ^{208}Pb. This indicates that the dynamics of the nuclear reaction towards the synthesis of the heaviest nuclei must be reconsidered.

Nuclear Structure

The experimental study of K isomeric states in the region of the heaviest nuclei is ongoing at TASCA. The analysis of the experimental data on the synthesis of the extremely short-lived ^{252}Rf via its anticipated long-lived K-isomeric state as suggested in [J. Khuyagbaatar, Eur. Phys. J. A 58, 243 (2022)] was finalized. The 2n channel of the ^{50}Ti+^{204}Pb reaction was used to produce this isotope. The presence of a µs-fission activity was observed at beam energies

corresponding to the 2n channel. However, for an unambiguous conclusion on the discovery of the new isotope ^{252}Rf, additional data are needed, which are foreseen to be collected in 2024.

The analyses of the experimental data on the fission-fragment mass distributions of ^{252}No, ^{255}Rf and ^{258}Rf measured with the ANSWERS setup in the FAIR Phase-0 beamtimes in 2020-2022 were finalized and will be published. The analyses of the experimental data on the low-lying structure of ^{253}No and ^{255}No were finalized as well and will also be published.

At SHIPTRAP, the buffer-gas cell was extensively tested with the use of radioactive recoil sources to understand and overcome the change in the entrance window thickness due to deposition of contaminants from the beamline during its extended operation online at cryogenic temperatures. A heating system was added closely to the entrance window to ensure the evaporation of the residues from its surface and to maintain a constant stopping efficiency in preparation of the beamtime scheduled in 2024. A systematic optimization study of the mass resolving power of the preparation trap has been performed and different settings for fast/slow ion bunch centering and purification have been established.

The data acquisition software has been updated with a new reconstruction algorithm, which allows recovering incomplete events from the position-sensitive ion detector. The new software is Python-based and is intended to substitute the existing Labview solution, in line with the GSI general migration away from National Instrument software products. A database has been established for a more efficient data storage of all the environmental parameters that must be kept under control during long measurement times.

The analysis of the SHIPTRAP data from the beamtime 2021 has progressed, and specific libraries have been customized. The data analysis of the measurement of ^{257}Rf, as well as of the α-decay chain ^{206}Fr – ^{202}At – ^{198}Bi were finalized. A deeper understanding of some systematic uncertainties requires additional offline measurements. Publications are in preparation.

Atomic Physics

As there was no physics beamtime at GSI in 2023, the program on the laser spectroscopic investigations of the heaviest elements focused on the analysis of the results from the experimental campaigns of the preceding years. The results of 6 on-line produced fermium (Fm, Z=100) isotopes obtained with the RADRIS technique in the FAIR Phase-0 beamtimes in 2020-2022 were evaluated and combined with results on two more fermium isotopes, which were measured off-line at JGU Mainz. The experimental results were complemented by state-of-the-art nuclear model calculations and a comprehensive manuscript reporting on these findings was submitted in 2023. The technical developments and results on probing the opportunities and limitations of in-gas-cell laser spectroscopy of the heaviest elements with RADRIS were published as a proceedings contribution to the EMIS2022 conference [2]. In the beamtime 2022 the new JetRIS setup was commissioned. Here, laser spectroscopy was performed in an effusing gas-jet to improve the spectral resolution, which could be demonstrated on-line with a measurement of the nobelium isotope ^{254}No. Here, a discrepancy to the transition energies with respect to the earlier RADRIS measurements was found, which demanded additional systematic investigations of the used wavemeter to quantify all systematic effects in the photon energy determination. These results were finalized and the manuscript on the in-gas-jet laser spectroscopy of ^{254}No with JetRIS was submitted in 2023. As the efficiency of the setup was found to be not ideal, further investigations with the setup were performed in 2023 to improve the setup and to understand the main sources of losses. An improvement in the efficiency by a factor up to 2 was achieved and the behavior was compared to finite-element computer simulations. This optimization process is still on-going and quite promising that a significantly improved setup is available for the upcoming beamtime, which is scheduled for 2025. Further developments, which took place at the HI Mainz, were performed with a new quadrupole mass spectrometer setup to test and evaluate ionization schemes for later on-line use as well as with the assembly and testing of a new multi-reflection time-of-flight mass spectrometer. The latter will in future extend the capabilities of the group's gas-cell laser spectroscopy program to long-lived nuclides and to nuclides independent of their respective decay mode.

The collaboration with the institute of physics and the department of chemistry at JGU Mainz, which enables measurements of long-lived actinide isotopes with minuscule sample sizes at the RISIKO mass separator, continued also in 2023. The data from previous measurement campaigns yielding nuclear moments and isotope shifts of the actinide isotopes $^{249-253}$Cf probed by laser spectroscopy was published [3]. A new measurement campaign that aims at probing the atomic and nuclear properties of ^{255}Fm was performed in 2023. For this, an ^{254}Es sample from ORNL Oak Ridge, TN, USA was shipped to the high-flux reactor at the ILL Grenoble, France, to breed ^{255}Es ($T_{1/2}$: 40 d) which decays into ^{255}Fm ($T_{1/2}$: 20 h). Using chemical separation techniques, 17 samples of ^{255}Fm became available

over the course of about three months. Some of these samples were used for measurements on the atomic fine and hyperfine structure; the data are under analysis. Other samples were used for collaborative work with the GSI Biophysics department, cf. section "Technical developments and key contributions to collaborative work".

Chemical Studies

Building up on the success of experiments conducted during the FAIR Phase-0 beamtime in 2022, our research expanded to explore the properties of Hg, Po, and At isotopes, serving as lighter homologs of the superheavy elements Cn, Lv, and Ts. Collaborating with the CTU Prague from FAIR aspirant partner Czech Republic, we conducted gas-phase chromatography experiments with gamma-decaying isotopes of Hg, At (as homolog of Ts), and Po (as homolog of Lv) at NPI CAS Řež (CZ). We employed a new and versatile setup, designed to study the interaction of Hg, At, and Po with quartz surfaces. The temperatures of the chromatography column ranged from +1000 °C to -55 °C in thermochromatography (TC) studies, and from +350 °C to +20 °C in isothermal chromatography (IC) experiments. The radioisotopes were produced in fusion-evaporation reactions using a 48-MeV ^{3}He-beam, recoiling from the thin target, and thermalized in helium gas. This also served as a carrier gas to transport the volatile At and Hg to the column. Po isotopes were collected in a Ti catcher foil placed directly behind the target during irradiation. After the end of irradiation, the foil was placed in the chromatography column and heated to release the collected Po isotopes. Reactive gases, such as oxygen and hydrogen, could be introduced directly before the chromatography column. In IC experiments with At, the fraction of radioisotopes surviving the transport through the chromatography column was deposited in a charcoal filter measured using a gamma-detector. The experiments revealed a complex interaction of At with the quartz surface. In TC experiments, yielding internal chromatograms, Hg was found not to be adsorbed on quartz, whereas it reacted strongly with Au, which agrees with known data [L. Lens et al., Radiochim. Acta 106 (2018) 949]. The data on At and Po are under final analysis. For these experiments, the MARGE subsystem [4] for production and delivery of radionuclides was extended to house two independent recoil target chambers, which were utilized for two simultaneous and independent experiments using aqueous chemistry and gaseous chemistry in parallel during the same beamtime. Supported by the FAIR-CZ national project, development of an on-line microfluidic aqueous chemistry apparatus for the NuSTAR collaboration continued. As a part of the testing, the early studies of chemical properties of the Sg and Nh homologues were extended; the results are in the final stage of analysis and are planned to be published.

First gas chromatography studies with the short-lived 145-ms isotope ^{216}Po were conducted offline at GSI. The experiments were performed offline using a ^{224}Ra source placed inside the TASCA Recoil Transfer Chamber, to which a miniCOMPACT detection system was attached. The measured chromatograms of ^{216}Po in helium, argon, and oxygen carrier gases point at a rather low volatility of Po species over the silicon oxide surface kept at room temperature.

Chemical Theory Supporting Experimental Work

To assist current gas-phase chemistry experiments on the volatility of At, a homolog of Ts, and to predict the behaviour of Ts in future experiments, calculations of adsorption energies E_{ads} of these elements and their compounds on gold and hydroxylated quartz surfaces were performed using relativistic periodic density functional theory implemented in the AMS BAND software. The following compounds were considered: MO, MH, MO_2, OMO, MOH, MOO, OM(OH) and MO(OH), where M = At and Ts. The obtained values of E_{ads} indicate that all the molecules should interact fairly strongly with the gold surface, with those of Ts being more reactive than the At ones. The similarity of the E_{ads} values of all the considered At compounds will make it challenging to differentiate between them via measurements of their adsorption enthalpies, given experimental uncertainty. However, the differences in E_{ads} among Ts compounds are more pronounced, so that one should be able to differentiate between the species.

Results for the adsorption of At and Ts on the hydroxylated quartz surfaces have shown that elemental At should adsorb very weakly, with E_{ads} of -26 kJ/mol on geminal and -20 kJ/mol on vicinal silanols, while AtH, AtO, AtO_2, and AtOH should adsorb more strongly, with E_{ads} of -30 to -40 kJ/mol. The E_{ads} absolute values of OAtOH, AtO(OH) and OAtO are the largest, reaching 100 kJ/mol. Thus, it should be easy to distinguish between adsorption of elemental At and that of its compounds on the quartz surface, i.e., elemental At should be much more volatile. The corresponding Ts compounds should be more reactive than those of At, i.e., they should adsorb at higher temperatures, than those of At. It should therefore be possible to distinguish between the At and Ts species. Also, the differences in E_{ads} between the various species of Ts are larger than those between the At ones.

Adsorption properties of group 1 and 2 elements and their compounds including those of elements 119 and 120 on hydroxylated quartz surfaces were calculated using a periodic BAND suite [5]. The results show that all the considered group 1 and 2 elements should adsorb rather moderately on the quartz surfaces, with E119 and E120 most weakly, due to the strong relativistic stabilization and contraction of the 8s atomic orbital. This means that E119 and E120 should have a deposition peak in the quartz chromatography column with a temperature gradient from room temperature to far below zero in the sequence Cs/Ba > Fr/Ra > E119/E120. For group-1 element MH and MOH molecules, the adsorption energies are high, so that the adsorption-desorption equilibrium should be reached at very high temperatures, with the following trend in the adsorption strength MH > MOH >> M.

Optimised Norm-Conserving Vanderbilt Pseudopotentials for Actinides and Super Heavy Elements in the PseudoDojo have been developed in a large collaboration with other theory groups. Our work comprised calculations of solid-state structures of the elements using the BAND software. The new approach should allow calculations of solid-state properties of superheavy elements using, e.g., the Quantum Espresso software, at a higher level of theory than presently available.

Technical developments and key contributions to collaborative work

As discussed in the section “Atomic physics”, 17 samples of ^{255}Fm became available over the course of about three months. Five of these samples were used for collaborative work with the GSI Biophysics department to test the uptake and cytotoxicity of ^{255}Fm in prostate cancer cells. For this purpose, five prostate specific membrane antigen (PSMA)-617 samples were labelled with ^{255}Fm; PSMA-617 is a small molecule which binds with high affinity to the transmembrane glutamate carboxypeptidase PSMA that is highly expressed on prostate cancer cells, such as the PC3-PIP cells that were used in this study. Target-specific uptake of ^{255}Fm-PSMA was evaluated by using liquid scintillation counting (LSC) and the effect of the single alpha-emitter ^{255}Fm on PC3-PIP cell proliferation was assessed with the colorimetric MTS assay, confirming a strong dose-dependent decrease in cell viability.

Further work at HI Mainz and JGU involved the development of laser resonance chromatography (LRC) to investigate the atomic structure of superheavy elements [M. Laatiaoui et al., Phys. Rev. Lett.125, 023002 (2020)]. The LRC apparatus is now in operation. The chromatographic performance of the apparatus was evaluated by analyzing the arrival time distributions (ATDs) of laser ablated Hf^+ ions and the ATD peak separation when comparing Lu^+ and Yb^+ ions in their ground states. A metastable ATD peak was observed for the first time in the Lu^+ arrival time distributions. The LRC was also successfully demonstrated for the first time by initiating the optical $^1S_0$-$^3P_1$ ground state transition in this ion at about 28,503 cm^{-1}, allowing optical pumping to the metastable $^3D_1$ state. We measured the hyperfine parameters of the $^3P_1$ state in $^{176}Lu^+$ and determined the isotopic shift of the spectral line relative to that of the more abundant $^{175}Lu^+$. To measure the extraction and transmission efficiencies, $^{219}Rn^+$ recoil ions from a ^{223}Ra source were used. In a typical bunching operation, the overall efficiency of the device was found to be 0.6 %. Before conducting future studies at in-flight separator facilities, the LRC technique needs to be further optimized to investigate the spectral precision of the method and improve the overall efficiency of the apparatus. Further efforts have been made to investigate the transport properties of heavy metal ions in buffer gas environments. The studies complement the LRC investigations and provide a deeper understanding of the underlying ion-atom interactions. For this purpose, a Cryogenic Ion Mobility Spectrometer (CIMS) was designed, developed, and recently put into operation. Systematic investigations of ion mobility in a wide range of reduced electric fields were carried out for some lanthanides and transition metals, and metastable states were observed for some of them for the first time. This research will be extended to actinide cations in the future.

At HI Mainz and in collaboration with the GSI Materials Research department, work is ongoing towards the production of improved f-element targets for accelerator experiments, optimized for coping with highest beam intensities as they will become available, e.g., with HELIAC. Novel developments in electrochemistry were transferred to target production, employing anhydrous electrochemical routes. The produced thin layers were characterized by a variety of analytical methods, irradiated with ^{48}Ca ions at TASCA/X8 and with ^{197}Au ions at M3, and characterized again after irradiation. The study is published in [6].

Also at HI Mainz, the production of tailor-made samples of exotic radionuclides continued to be an important pillar of the SHE Chemistry program. Besides the ^{255}Fm samples described above, optimized samples of ^{242}Pu for the production of the fission isomers in ^{240,242}Am at the IGISOL facility at U. Jyväskylä (SF) were delivered and successfully used in an accelerator experiment at Jyväskylä. Samples of ^{232}Th were prepared for collaborative work with U. Gothenburg and U. Stockholm for experiments at the Stockholm DESIREE storage ring. Studies on the formation of few-atom clusters of thorium and uranium were carried out in collaboration with U. Greifswald.

The chemical study of elements beyond Mc requires the development of novel techniques to efficiently transfer short-lived isotopes with half-lives below 100 ms to a gas chromatography detector array. The proposed universal buffer gas stopping cell (UniCell) [V. Varentsov et al., Nucl. Instrum. Meth. A 940 (2019) 206] is based on the radiofrequency (RF) ion-funnel technique and is designed to succeed the TASCA recoil transfer chamber. Ion trajectory simulations for UniCell were carried out using SIMION. A setup with DC field strength E = 100 V/cm, temperature T = 300 K, helium pressure P = 1 bar, and peak-to-peak RF amplitude V_{pp} = 200 V was found to be desirable as the optimum choice for an extraction efficiency of 100%. The extraction time was calculated to be about 4.4 ms and 2.2 ms for ions of mass 293 amu and charge states 1+ and 2+, respectively. The fabrication of the UniCell RF-funnel assembly consisting of 350 electrodes was performed by our collaborators at ITE Cracow, Poland. Mechanical design on its integration and the development of suitable electronics have commenced. First tests with radioactive sources to start the commissioning of the device are being prepared. In the next step, the offline test of UniCell will be carried out to benchmark the simulation results. In addition, the high-pressure Ion Transfer by Gas Flow (ITGF) device to couple UniCell to the COMPACT detection setup was proposed and initially studied using COMSOL Multiphysics®. The ITGF transport time decreases with increasing gas flow rate. After optimizations of the ITGF, the flow rate is selected to be more than 20 mL/s, and the time to pass through the ITGF device is only about 0.2 ms. The results of the above simulations are in preparation for publication.

The focal-plane area of the TASCA separator (cave X8) was reconstructed in preparation for the ANSWERS spectroscopy beam time in 2025. The whole area where the ANSWERS setup is installed was surrounded by shielding that resembles a hut with a movable door. The effect of this extra shielding on the neutron-background was tested during the engineering run in November 2023 together with the Radiation Safety Department. The result demonstrated a substantial decrease (about a factor of ten) in the neutron-background.

In the process of replacing the 40-year-old SHIP magnet power supplies, new power supplies for the quadrupole magnets were delivered and installed. This process was performed together with the GAT, EPS and ACO groups of GSI. The functionality of the new power supplies was tested in the engineering run in November 2023. Here, one day of ^{40}Ar beam was used with ^{169}Tm and ^{208}Pb targets, The obtained results of the fusion products in rate and spatial distribution match the expectations and thus SHIP is again ready for the upcoming physics beamtime 2024. The dipole magnet power supplies are next to be exchanged.

Outlook for 2024

At SHIP, a main beamtime with ^{40}Ar and ^{50}Ti beam to perform mass measurements of the ground and isomeric state of ^{258}Db and lighter Fr, At, and Bi isotopes is scheduled for SHIPTRAP in 2024. The experiment aims at disentangling the ground state from the low-lying long-lived metastable state and at accurately determining its excitation energy. Taking into account the half-lives of these two states, the tentative identification of the ground and isomeric state in ^{258}Db provided by previous decay spectroscopy studies at SHIP [M. Vostinar et al., Eur. Phys. J. A 55, 17 (2019)] can be verified. However, this measurement is quite challenging as the count rate of ^{258}Db is of the order of 1 ion every 5 hours at the SHIPTRAP position-sensitive detector. A second, parasitic beam time with chromium beam is planned at SHIP and is scheduled for May 2024; it will be devoted to laser spectroscopic studies with lutetium isotopes, the iso-electronic homologue of $_{103}$Lr. Here the desorption behaviour with the RADRIS technique will be evaluated to understand the expected behaviour of Lr, for which an extended level search is foreseen in 2025.

At TASCA, chemistry studies towards seaborgium carbonyl complex formation and its reactivity, volatility, and the chemical stability are planned for 2024. The newly tested combined detection system, miniCOMPACT plus COMPACT, allows for studies of carbonyl complexes with very short-lived isotopes of the superheavy elements, which can be produced in cold fusion reactions for elements up to $_{107}$Bh, with larger production rates than more long-lived isotopes from hot fusion reactions. This will open the perspective for the first study with carbonyl complexes of Bh, which are unknown yet.

The chemistry studies at NPI CAS Řež will continue with another beamtime in spring 2024 to further the understanding of the properties of Hg, Po, and At in contact with quartz surfaces, and the offline studies with ^{216}Po will be extended to cover a wider temperature range. These will be accompanied by theoretical work on the volatility of Po, a homolog of Lv, yielding predictions of the adsorption behavior of these elements and their compounds on surfaces of gold and quartz on the basis of the relativistic periodic DFT calculations. Thermodynamic properties of SHEs as a function of temperature are planned to be investigated as well.

Selected publications of 2023

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