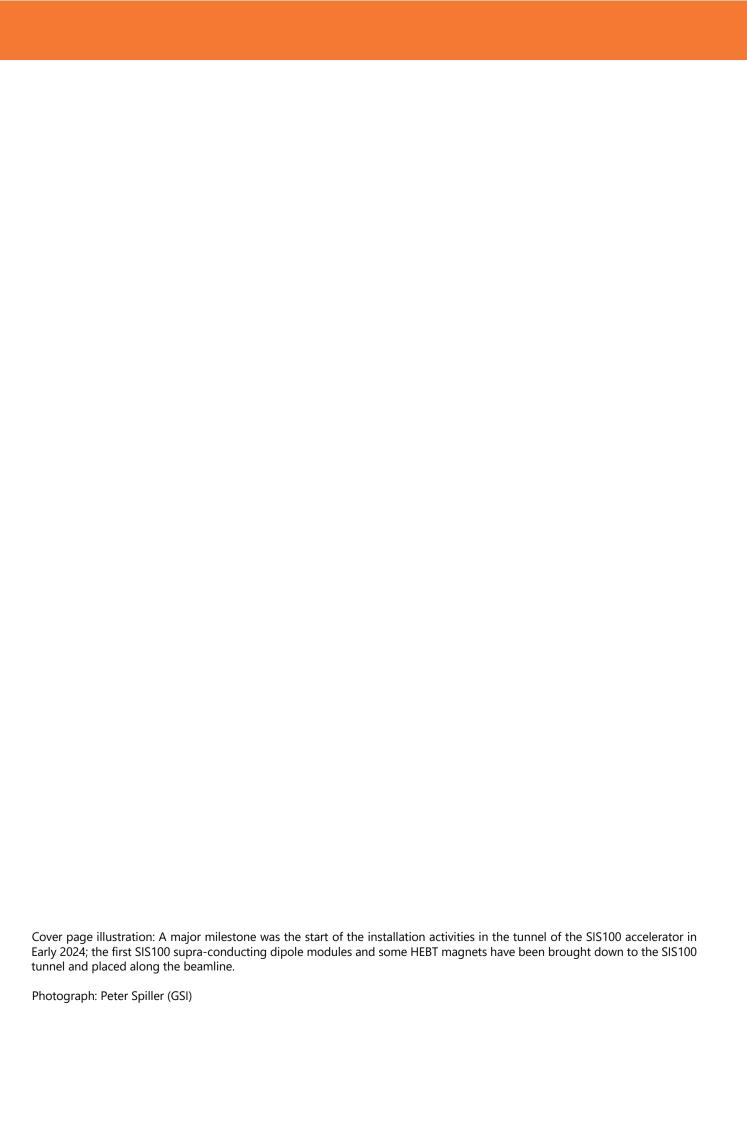
GSI-FAIR SCIENTIFIC REPORT 2024

An overview of the 2024 achievements in science and technology









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4.5 Superheavy elements at GSI and HI Mainz

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A meta-analysis of nuclear data confirming the existence of a region of increased nuclear stability in the neutron-rich region of the heaviest elements was extended by comparing predictions from a variety of nuclear models. These support the conclusion of the existence of an island of enhanced stability, but make clear that the location of the peak (i.e., the longest-lived nucleus) and the extension of the island are currently not reliably predicted. On the experimental side, the focus in 2024 was on running experiments within the user beamtime served at GSI; additional activities at GSI focused on the analysis and publication of data obtained previously, both at GSI as well as abroad, e.g., at NPI CAS Řež (CZ). Online chemistry studies were performed at NPI CAS Řež (CZ). Furthermore, 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 ²⁵⁵Fm obtained from a 40-d ²⁵⁵Es 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 [1]. 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.

Synthesis / Nuclear Reactions

The discovery of superheavy elements (SHE) beyond Og (Z=118) remains as a hot topic in the field. Since first attempts to synthesize the elements 119 and 120 in the ⁵⁰Ti+Bk/Cf reactions at the gas-filled recoil separator TASCA [2], intensive nuclear reaction studies were carried out in collaboration with the Australian National University (ANU), Canberra, Australia, at the Heavy Ion Accelerator Facility of ANU. The projectile ⁵⁰Ti has been shown to be most promising for the synthesis of elements beyond Og [3], which confirmed the choice for the most promising reaction for the syntheses of 119/120 at TASCA. On the other hand, one of the unresolved issues is the question of how much the cross section will be reduced for ⁵⁰Ti-induced reactions with deformed targets compared to the known ⁴⁸Ca reactions. To assist for this problem, the cross sections of three different types of fusion-evaporation reactions with different entrance channel properties were analyzed [4]. For such a compilation, it was necessary to measure the cross sections for ⁵⁰Ti- and ⁴⁸Ca induced reactions with deformed targets, which were made at TASCA. The results show the Coulomb force to be the main issue for fusion of the heavy nuclei by substantially increasing the probability of processes preceding fusion, such as quasi-fission. Based on these data and the new systematics, the maximum cross sections for various types of reactions leading to SHN were predicted including the elements 119/120. The recent experimental data for ⁵⁰Ti+²⁴⁴Pu and ⁵⁴Cr+²³⁸U reactions measured at the Berkeley Gas-filled Separator at LBNL Berkeley, USA and the Dubna Gas-filled Recoil Separator at FLNR, JINR, Dubna, Russia, respectively, are in satisfying agreement with the predictions. A continuation of the started ⁵⁰Ti+²⁴⁹Cf experiment at TASCA would thus appear sensible.

Nuclear Structure

At TASCA, the nuclear structure research program with the focal plane detection system is ongoing. The search for the extremely short-lived ²⁵²Rf was completed in 2024 and the results are now published [5]. This isotope was discovered in the 2n channel of the ⁵⁰Ti+²⁰⁴Pb reaction by using its long-lived K-isomeric state. The K-isomeric state with a half-life of ≈13 µs enabled the separation of ^{252m}Rf at TASCA during its flight time of about 1 µs. Once implanted, ^{252m}Rf decays via an electromagnetic transition to the ground state; the fast digital electronics was able to register the conversion electrons emitted in this decay. The ground state then undergoes fission with a half-life of ≈60 ns, as deduced from three observed events. This very short value expands the range of half-lives of the known superheavy nuclei by about two orders of magnitude. Our findings set a new benchmark for further exploration of phenomena associated with high-K states and inverted fission-stability in the heaviest nuclei. The electron-capture delayed fission (EDCF) from 242Es has been revisited as well, by producing this nucleus in the 48 Ca+ 197 Au reaction. The probability of EDCF was measured directly and with improved statistics [6]. In separate work, the most probable outcome of ternary fission, which is the emission of two heavy fragments and one light charged particle, has been studied. In about 90% of all cases, these are α particles, which are often referred to as long-range alpha (LRA). Such decays have been extensively studied over decades in various heavy fissioning systems and the absolute probability for these processes was found to be about (0.2-0.4) % relative to binary fission. A possible occurrence of this process in superheavy nuclei was considered in recent theoretical work [7]. As a result, it is not excluded that the probabilities of LRA emission are substantial (up to the percent level) in the fission of neutron-deficient heavy and superheavy nuclei.

At SHIPTRAP, a compact buffer-gas cell was designed to enable the use of radioactive recoil sources for offline mass measurements. This mainly concerns heavy nuclides, in particular long-lived actinide isotopes that can be obtained in macroscopic amounts. Such ions can serve as reference ions in online mass measurements and allow us to track changes of the magnetic field of the SHIPTRAP solenoid magnet.

Atomic Physics

The results of laser spectroscopy 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 findings along with theoretical calculations were published in 2024 in Nature [8]. For the JETRIS setup, where the laser spectroscopy is performed in an effusing gas-jet to improve the spectral resolution, the results from the commissioning beamtime 2022 of the in-gas-jet laser spectroscopy of ²⁵⁴No were finally published in 2024 [9]. The investigation and optimization of the JetRIS setup was continued with the help of collaboration partners from KU Leuven, who contributed to the design and testing of new geometries which led to a promising improvement in the setup off-line efficiency. These improvements will allow probing the 8⁻ K-isomer in ²⁵⁴No in the next beamtime, which is scheduled for February 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. For the laser spectroscopy program, a parasitic beamtime with a 52Cr beam was performed in May 2024. Using a 107Ag target the alpha-decaying isotopes 155,156Lu were produced. Lutetium is the iso-electronic homologue of 103Lr and these investigations were planned to benchmark the on-going Lr level search. Unfortunately, this effort was hampered by the fact that mainly the short-lived isomeric state 156m Lu ($T_{1/2}$ =198 ms) was produced in this reaction. This short lifetime severely reduces the effective efficiency of the cyclic operation of the in-gas-cell laser spectroscopy with the RADRIS setup. In summary, there was no evidence for laser ionization of lutetium and the data is still under evaluation to understand this behavior. Besides the study of lutetium isotopes, the neutron-deficient isotopes ¹⁵²Tm and ¹⁵¹Er were investigated by laser spectroscopy for the first time. Here the data is under analysis with the aim to extract nuclear properties on the change of the mean square charge radii.

The collaboration with the Institute of Physics and the Department of Chemistry at JGU Mainz enabled measurements of long-lived actinide isotopes with minuscule sample sizes at the RISIKO mass separator in 2023. The data on ²⁵⁴Cf and on the atomic structure and the hyperfine structure in Fm isotopes was continued to be analyzed. In the latter, additional input was obtained from theoretical atomic calculations by J. Andrews, which helped in understanding the obtained data. At the current stage, the manuscripts are in preparation for publication in 2025.

Chemical Studies

At TASCA, chemistry studies on seaborgium carbonyl complex formation and its reactivity, volatility, and the chemical stability were performed in 2024. The newly tested combined detection system, miniCOMPACT plus COMPACT, allowed 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 ¹⁰⁷Bh, with larger production rates than more long-lived isotopes from hot-fusion reactions. To verify this approach, a study with carbonyl complexes of ¹⁰⁶Sg was performed at TASCA. The isotope ²⁵⁹Sg was produced via the nuclear reaction ⁵²Cr + ²⁰⁸Pb and pre-separated with TASCA. The Sg recoils were thermalized in a gas mixture of helium and carbon monoxide (CO). More than 60 decay chains originating from ²⁵⁹Sg were registered in the combined detection setup. The Sghexacarbonyl complex is formed in the reaction of Sg ions/atoms with CO ligands via a multi-step process. The intermediate reaction products are more reactive and non-volatile, while the final product Sg(CO)₆ shows a low interaction strength with a detector surface and adsorbs at a low temperature by physisorption. This study opens the perspective for the first study with carbonyl complexes of Bh, which are yet unknown.

Building on the success of experiments conducted in 2023 in collaboration with the CTU Prague from FAIR aspirant partner Czech Republic, we conducted gas-phase chromatography experiments with gamma-decaying isotopes of Hg (as homolog of Cn), Tl (as homolog of Nh), Po (as homolog of Lv) and At (as homolog of Ts) at NPI CAS Řež (CZ). We employed a versatile setup, designed to study the interaction of Hg, Tl, Po and At with quartz and alpha-Al₂O₃ surfaces of different chemical reactivities. The temperature gradients of the chromatography column ranged from +1000°C to -170°C in thermochromatography studies. The radioisotopes were produced in fusion-evaporation reactions using a 48-MeV ³He-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. The non-volatile Po and Tl isotopes were collected in a Ti, Ta or C 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 or Tl isotopes. Reactive gases, such as oxygen or water vapor could be introduced directly before the chromatography column. The experiments allowed the determination of the adsorption enthalpy of elemental polonium on quartz and alpha-Al₂O₃ surfaces. Furthermore, elemental At was deposited at temperatures below -60°C on quartz and the complex interaction of Tl, At, and Po compounds with the quartz surface was investigated. Hg was adsorbed on quartz at -130°C, which agrees with known data [10]. The data on Hg, Tl, Po and At are under final analysis.

Chemical Theory Supporting Experimental Work

To assist current gas-phase chemistry experiments on the volatility of At and Po, homologs of Ts and Lv, respectively, and to predict the behaviour of Ts and Lv in future experiments, calculations of formation reaction energies and 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. For adsorption on gold, for group 15, the compounds under investigation were MH₃ and MO(OH), where M = Bi or Mc, in addition to the previously considered M and MH. For group 16, the MO, MO₂ and MH₂ molecules, where M = Po and Lv, were considered, in addition to the previously considered M and MH.

The results have shown that for group 15, the compounds of Bi and Mc should be rather distinguishable by their adsorption on gold. The sequence in the adsorption strength should be MOOH > M > MH > MH₃, with the Mc species being less strongly adsorbed.

For group 16, the MH_2 compounds of Po and Lv are predicted to be the most volatile over gold among the considered ones. The adsorption temperature should be slightly higher than room temperature, with the adsorption of other compounds occurring at considerably higher temperatures. The PoO, PoH and PoO₂ molecules have nearly identical adsorption energies, making them indistinguishable from each another in experimental settings. This is also the case for the following pairs of molecules: LvO and LvH. Calculated E_{ads} of Po and PoO₂ are in very good agreement with the experimental ΔH_{ads} data for these species confirming the experimental observation stating that "PoO₂ has a lower affinity for gold compared to polonium". The claimed BiPo should have not been observed at slightly lower - ΔH_{ads} values, however, this supposition should be further checked.

According to the results, in comparison with Po, LvH and LvO should be deposited at lower adsorption temperature, while it should be the other way round for MO_2 and MH_2 compounds. It should be possible to differentiate between Po and Lv by adsorption of these elements and their species on the gold surface.

We have also started the study on adsorption of Po and Lv on hydroxylated quartz surfaces. Considered species are M, MH₂ MO and MO₂. Preliminary results for geminal and vicinal silanols show that the elemental Po should be very volatile over quartz, followed by PoH₂, PoO, and PoO₂. The work is still in progress for Lv and other types of modified quartz surfaces.

Technical developments and key contributions to collaborative work

Further work at HI Mainz and JGU involved the development of laser resonance chromatography (LRC) to investigate the atomic structure of superheavy elements [11]. 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 1S₀ - 3P₁ ground state transition in this ion at about 28,503 cm⁻¹, allowing optical pumping to the metastable ³D₁ state. We measured the hyperfine parameters of the ³P₁ state in ¹⁷⁶Lu⁺ and determined the isotopic shift of the spectral line relative to that of the more abundant ¹⁷⁵Lu⁺. To measure the extraction and transmission efficiencies, ²¹⁹Rn⁺ recoil ions from a ²²³Ra source, were used. In a typical bunching operation, the overall efficiency of the device was found to be 0.6 %. First commissioning results were published in [12]. 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.

Also at HI Mainz, the production of tailor-made samples of exotic radionuclides continued to be an important pillar of the SHE Chemistry program. A ²³¹Pa sample was prepared for ISOLDE @ CERN. The sample was used for systematic studies of protactinium molecules (CERN ISOLDE LOI-258). A legacy ²³¹Pa solution from the stocks at Nuclear Chemistry in Mainz was used, which was purified by column chromatography. Subsequently, 1 kBq of the purified solution was dripped into a Ta container provided by CERN, dried, shipped to CERN and used for the production of Pa beams. High-precision mass measurements of artificial ¹⁶³Ho, and its electron-capture decay daughter ¹⁶³Dy have been performed at PENTATRAP at MPIK Heidelberg; this work assists the quest for the determination of the mass of the electron neutrino by providing an independent highly precise Q-value of this reaction [13].

The chemical study of elements beyond Mc requires the development of novel techniques to efficiently transfer short-lived (tens of ms) isotopes with half-lives below 100 ms to a gas chromatography detector array. The proposed universal buffer gas stopping cell (UniCell) [14] is based on the radiofrequency (RF) ion-funnel technique and is designed to succeed the TASCA recoil transfer chamber. Ion trajectory and gas-dynamic simulations have been finalized and the submitted publication is now under review. The advanced ion funnel with an electrode spacing of only ca. 0.1 mm was fabricated by ITE Cracow and arrived at GSI. First electronic circuits to provide the required RF signal to the device were built but do not yet reach the required voltages for operation. To improve the performance, four interfacing printed circuit boards to adapt the 350 electrical contacts of the funnel have been designed in collaboration with the GSI experiment electronics department to match the exact dimensions. After finalizing the electronics and ongoing mechanical works, the commissioning with short-lived alpha-emitting isotopes is planned.

The process of replacing the 40-year-old SHIP magnet power supplies was continued by the procurements of new power supplies for the dipole magnets which were delivered and installed in December 2024. This process was performed together with the GAT and ACO groups of GSI. The functionality of the new power supplies will be finally tested and integrated into the GSI accelerator control system with the aim to use the new power supplies for the beamtime which starts in February 2025. The new dipole power supplies are already within the FESA framework of the new control system of the GSI accelerator chain. After the beamtime the extended break resulting from the ongoing renovation of the experimental hall, will be used to migrate the SHIP control to the new controls system.

Outlook 2025

For the laser spectroscopy program two main beamtimes are scheduled for 2025. The beamtime in February 2025 will be devoted to the in-gas-jet laser ionization with the JETRIS setup. The main goal is the measurement of the Hyperfine structure splitting of the 8^- - K^{π} -isomer in 254 No. As the nuclear g-factors result from single particle properties, e.g., specific nuclear orbitals, a measurement of the nuclear magnetic dipole moment will enable an assignment of the constituents of the nuclear configuration in this particular isomer. In the second beamtime in June 2025 measurements with the in-gas-cell setup RADRIS will be conducted, Especially the, the long-lived isotope 246 Cf , along with other Cf-isotopes will be investigated using a new detector setup consisting of 8 individual detectors to observe the laser excitation signal for different wavelengths on different detectors.

At TASCA, the ⁴⁸Ca+²⁴³Am reaction is planned to be used in Spring/Summer 2025 for studies of ²⁸⁸Mc and its decay products with the new ANSWERS setup.

Building on the success of chemistry studies with seaborgium carbonyl complex, the first study with carbonyl complexes of Bh, which are unknown yet, is proposed as the next chemistry experiment at TASCA. The results of Sg carbonyl study have demonstrated that the combined detection system, miniCOMPACT plus COMPACT, will allow for studies of Bh carbonyl complexes with very short-lived isotope ²⁶²Bh, which can be produced in cold fusion reaction ⁵⁵Mn + ²⁰⁸Pb using the newly developed ⁵⁵Mn ion beam from the PIG source.

The chemistry studies at NPI CAS Řež will continue to further the understanding of the properties of Hg, Po, and At in contact with quartz surfaces, and the offline studies with ²¹⁶Po 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 and gold and quartz on the basis of the relativistic periodic DFT calculations.

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