4.4 Superheavy elements at GSI and HI Mainz

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In 2021, activities at GSI focused on the UNILAC beamtime within the FAIR Phase-0 program. These comprised chemistry studies of Nh, high-precision mass measurements of Rf-Db, on-line commissioning of the novel ANSWERS setup, and laser spectroscopy of Fm isotopes. In addition, the analysis of data obtained in previous beamtimes was continued. At HIM, the advancement of technical and methodological developments was most central, for example for applications in laser spectroscopy and mass spectrometry as well as radionuclide layer production for various applications. In addition, preparations for the beamtime 2022 have been performed.

Highlights in 2021

Synthesis

To further our understanding of nuclear reactions, especially on the dynamics of nuclear fusion reactions and effects on the de-excitation of compound nuclei, collaborative experimental work of the SHE-Chemistry department has been carried out at Australian National University's (ANU) Heavy Ion Accelerator Facility, Canberra, Australia [K. Banerjee et al., Phys. Lett. B 820 136601 (2021)] and at the Physics Department of Central University of Kerala, Kasaragod, India [A.C. Visakh et al., Phys. Rev. C 104, 054602 (2021)]. In fall of 2021, a one-week experiment on the study of multi-nucleon transfer reactions has been carried out at the MARA mass separator of University of Jyväskylä, Finland under the lead of the SHE-Chemistry Department (Spokesperson: J. Khuyagbaatar). The data analysis is ongoing.

Nuclear structure

At TASCA, nuclear structure experiments with the nominal large area focal plane detector are ongoing. By employing the sampling ADC "FEBEX" modules developed by the GSI's Experiment Electronics department, multi-isomeric states in ^{256}Rf have been successfully studied [J. Khuyagbaatar et al., Phys. Rev. C 103, 064303 (2021)]. Two isomeric states with half-lives of ≈14 and ≈10 μs have been observed with population rates of ≈18 and >10%, respectively. Such rates are typical for two-quasiparticle high-K isomeric states in this region of nuclei. Therefore, the observed two isomeric states have been attributed to have two-quasiparticle origin. Studying heavy nuclei for the presence of high-K isomeric states, especially those that have longer fission half-lives than their ground states (e.g., ^{250}No), demands to be continued.

Meanwhile, the ground state fission half-lives of neutron-deficient No and Rf isotopes are very short, coming very close to the present experimental limit of about one ms. Analysis of experimental data on the spontaneous fission half-lives of Rf isotopes in relation with their nuclear ground-state configuration hint at a potentially abrupt decrease in half-lives of unknown neutron-deficient Rf isotopes with neutron numbers <149. This suggests that no further neutron-deficient Rf isotopes exist. However, this conjecture was directly related to uncertainties in experimental data on ^{253}Rf. We revisited the decay of ^{253}Rf and identified two fission activities, which are attributed to decays of the two different states with half-lives of ≈12.8 ms and ≈44 μs [1]. In addition, the hitherto unknown α decay in ^{253}Rf, which is followed by α decay of the new isotope ^{249}No with a half-life of ≈15 ms, was observed. Based on our new data, no abrupt decrease in the half-lives of the neutron-deficient No and Rf isotopes is expected, in line with theoretical predictions. It is worth noting that the discovery of ^{249}No was also reported based on independent experiments performed at the SHELS separator at FLNR Dubna [A.I. Svirikhin et al., Phys. Part. Nucl. Lett. 18, 445 (2021)].

Presently, conclusive experimental data on the basic properties of superheavy nuclei are still scarce and are generally limited to data derived from α decay (energy, half-life) and spontaneous fission (half-life). This limitation is not only due to low production rates, but also because of limited detection efficiencies and signal isolation capabilities of the presently available experimental techniques, which do not allow extracting all information on the decay of superheavy nuclei. To solve this problem, the new Adsorption-based Nuclear Spectroscopy Without Evaporation Residue Signal (ANSWERS) setup was developed in the SHE Chemistry department. In 2021, in-beam commissioning of ANSWERS has been performed in the course of a one-week main beam (spokesperson: J. Khuyagbaatar), in which decays of Rf isotopes were investigated. As preliminary results of this beam time, we observed many multi-coincidence events between α particle, conversion electron(s) and photons associated with decay of ^{257}Rf. The observed data represent a completely new type of experimental data set, which was inaccessible with more traditional previously used setups. This required developing a new method of data analysis, and the data are currently under final analysis. At the same time, ANSWERS showed great potential for studies of fission of the heaviest nuclei, which is a long-term demanding topic that has not yet been satisfactorily explored.

In 2019 and 2020, flerovium isotopes were produced in two runs within FAIR Phase-0, using the fusion-evaporation reactions ^{48}Ca+^{242,244}Pu. They were studied with an upgraded TASISpec decay station positioned at the focal plane of the gas-filled separator TASCA, allowing for detailed nuclear spectroscopy of decay chains starting from ^{286,288}Fl [2] and ^{289}Fl. In 2021, details of the fifteen correlated α-decay chains starting from the odd-A superheavy nucleus ^{289}Fl were investigated. For this isotope, the global data set was roughly doubled. The results from the U310 nuclear spectroscopy experiment call for at least two parallel α-decay sequences starting from at least two different states of ^{289}Fl. Supported by α-electron and α-photon coincidences, α-decay fine structure could be established along the ^{289}Fl chain, and in particular for the α decay of ^{285}Cn, corroborated by Geant4 simulations. Two chains terminated by fast fission of ^{277}Hs, thereby confirming one earlier event observed at TASCA [Ch.E. Düllmann et al., Phys. Rev. Lett. 104, 252701 (2010)]. The observed fine-structure characteristics can be explained by a decay path through low-spin positive-parity states, based on extensive nuclear structure calculations employing the symmetry-conserving configuration mixing theory [J .L. Egido and A. Jungclaus, Phys. Rev. Lett. 125, 192504 (2020)]. The complexity of these "triaxial beyond mean-field" calculations demands about the same real time on multi-core clusters for each decay step and parity as the combined 3-weeks long experiment. Furthermore, previous, revised, and newly derived fission probabilities of even-even superheavy nuclei were compared with various theoretical predictions. The results on ^{289}Fl confirm the overarching picture of high-precision coincidence-spectroscopy measurements providing valuable anchor points for theoretical predictions on the voyage to the Island of Stability. For the future, a conceptually new decay station is being assembled in Lund, which aims to further enhance the quality and sensitivity of nuclear spectroscopy experiments.

The investigation of the heaviest elements by Penning-trap mass spectrometry with SHIPTRAP has been further extended during a successful beamtime in spring 2021. The emphasis was on the investigation of low-lying isomeric states in heavy nuclei that are so long-lived that their investigation by decay spectroscopy is difficult. The high mass resolving power of SHIPTRAP allowed us to determine the absolute excitation energies of these isomeric states accurately and unambiguously for the first time. A low-lying isomeric state in ^{241}Cf was expected to exist according to systematics in N=143 isotones that would predominantly de-excite via internal conversion with an estimated half-life of about 0.2 ms [J. Khuyagbaatar et al., Phys. Rev. C 102, 044312 (2020)]. We have observed this isomer for the first time even though our measurement time was more than one second. Thus, we conclude that the isomer's half-life exceeds the Weisskopf estimate by about three orders of magnitude [Table of Isotopes, edited by R. B. Firestone, 8th ed. (Wiley, New York, 1996)]. In the region of the superheavy elements, we have performed accurate measurements of the excitation energy of the 4.9 s-isomeric state in ^{257}Rf (mass resolving power up to 10^7). Also, the ground state masses of ^{257}Rf and ^{258}Db have been directly determined with high precision at count rates down to 1 ion/day. These achievements extend the knowledge on the evolution of the N=152 shell closure towards heavier nuclides based on Penning-trap mass spectrometry as a complementary tool to decay spectroscopy. In a parasitic experiment, the

interesting members of the α-decay chains ^{206}Fr - ^{202}At –^{198}Bi and ^{204}Fr – ^{200}At – ^{196}Bi were studied. In each of these nuclides two (low-lying) isomeric states, a 7^+ state and a 10^- state, were known from previous investigations of the hyperfine structure and mean-square charge radii [K. M. Lynch et al. Phys. Rev. C 93, 014319 (2016)]. However, the excitation energy of the lowest isomers was experimentally inaccessible and remained unknown. We have now determined this energy directly and with high precision for the first time, extending the previous studies. The analysis of the 2021 beamtime data is ongoing and systematic uncertainties of the method are determined in offline studies.

Atomic physics

The laser spectroscopic investigation of the heaviest actinide elements advanced further in 2021. In the FAIR Phase-0 beam time, we performed detailed laser spectroscopy of ^{249,254}Fm produced via the decay of ^{253,254}No as they are inaccessible in a direct production scheme. Here, the isotope shift of the 25 11^2 cm^{-1} transition was precisely determined, concluding the measurements started in the beam time in 2020. The data are presently under evaluation. In addition, we were able to improve the spectral resolution in the $^1S_1$-$^1P_0$ transition of ^{252}No that was previously studied [S. Raeder et al., Phys. Rev. Lett. 120, 232503 (2018)]. Technical developments focused on an improved operation cycle for the RADRIS method that relies on a collection and measurement cycle that hampers the investigation of short-lived nuclei. Employing fast high-voltage switches was proven to minimize losses in short cycles, enabling measurements of short-lived nuclides such as ^{251}No with a half-life of T½=0.8 s and improving the performance during the search for atomic levels in lawrencium that are planned for 2022. For the lawrencium level search an improved theoretical prediction was obtained [3].

For upcoming experiments on nobelium with an improved spectral resolution in laser spectroscopy a new gas jet setup was further commissioned at the HIM using stable isotopes. A thorough characterization of the nozzle shaping the supersonic gas jet was performed using images of the fluorescence from optically excited dysprosium atoms seeded to the gas jet. With these investigations the setup was optimized to spectral resolutions down to 230 MHz corresponding to a gas jet Mach number of M=6.5. These studies pave the way for a measurement of the $K=8^-$ isomer in ^{254}No in the 2022 beam time at GSI.

Additional off-line measurements at the RISIKO setup at the University of Mainz were performed in collaboration with the group of Prof. Klaus Wendt. These became possible in 2021 using a remaining ^{254}Es sample that was originally delivered from ORNL to Mainz in 2019 for laser spectroscopic investigations. This sample was post-irradiated at the high-flux reactor at ILL, Grenoble, France, to breed an increased amount of ^{255}Es ($T_{½}$=40 d). This isotope β^--decays to ^{255}Fm ($T_{½}$=20 h), which can be separated for laser spectroscopy studies. The irradiated sample was shipped back to Mainz just a few days after irradiation, where a separation of ^{255}Fm from its mother ^{255}Es was performed. Multiple Es and Fm samples were prepared for improved laser spectroscopic measurements. This was a unique chance to have access to both short-lived nuclides. In total four fermium separations were performed. Laser spectroscopy was performed on different optical transitions in ^{255}Fm with sample sizes ranging from $1{\cdot}10^6$ to $7{\cdot}10^6$ atoms of ^{255}Fm. Eventually, an improved measurement of the hyperfine structure of ^{255}Es with a better spectral resolution was achieved.

Off-line studies in preparation for a measurement of the hyperfine structure of the second lowest-lying nuclear isomer known to date, ^{235m}U, were carried out at University of Jyväskylä within the Laser Ionization and Spectroscopy of Actinides (LISA) MSCA-ITN. For this, several ^{239}Pu recoil ion sources with a total activity of 2.4 MBq were prepared by molecular plating from DMF solution and were characterized both in Mainz as well as in Jyväskylä. Reference studies will be performed with the even-even neighbour ^{236}U, which shows no hyperfine structure; for this, ^{240}Pu sources (about 940 kBq in total) were prepared. The radionuclides of interest will be studied by collinear laser spectroscopy, a technique that provides a high spectral resolution.

Atomic physics via ion-mobility studies

Another method to study the atomic structure has been recently proposed under the name of Laser Resonance Chromatography (LRC) [M. Laatiaoui et al., Phys. Rev. Lett. 125, 023002 (2020)]. This technique is compatible with superheavy element production and is designed to enable laser spectroscopy on lawrencium (Lr) and beyond. It uses laser excitations to change the ratio of ions in the excited metastable state to those in the ground state by optical resonance pumping. Because ions in different states have different transport properties in dilute gases, when they are injected into a drift tube with a constant electric field, they move at different velocities through the drift tube toward a particle detector, enabling state-specific ion separation and resonance detection.

The LRC setup is meanwhile almost complete, and the commissioning phase has already begun with the verification of the vacuum and the functionality of key components such as the buffer-gas stopping cell, the quadrupole mass filter, and the laser systems. The FPGA-based data acquisition system has been commissioned and tested along with the experiment control system. After this phase, testing of the cryogenic drift tube will initially begin with ion mobility measurements on laser-ablated Cu^+ ions.

On the theoretical side, Dirac-Coulomb Hamiltonian and multi-reference configuration interaction (MRCI) methods have been applied to predict the electronic structure of Rf^+ and Db^+ ions, the next two elements in focus after Lr^+. The results for Rf^+ have already been published [H. Ramanantoanina et al., Phys. Rev. A 104, 022813 (2021)]. The MRCI method has also been used to calculate the ion-neutral interaction potentials for the Lr^+-He and Rf^+-He systems. These potentials are important inputs for predicting the transport properties of these superheavy element cations. The results are very promising and will be published soon.

Chemical studies beyond copernicium: nihonium, flerovium, moscovium and perspectives for livermorium

Chemical studies of the superheavy elements were performed at TASCA and focused on Nh and Mc. They picked up on the successful TASCA beamtime in 2020, in which the $^{48}Ca+^{243}Am$ reaction was used to produce 0.17-s ^{288}Mc, which decays into 1-s ^{284}Nh, which was in the focus of these studies. Somewhat unexpectedly, also first signals associated with ^{288}Mc had been observed in 2020. Based on this and on an analysis of previous studies of Nh at FLNR Dubna, Russia and at TASCA [4], an improved setup with an advanced 64-element miniCOMPACT detector was built. In test experiments, a faster and more efficient extraction of short-lived chemically reactive species into the miniCOMPACT gas chromatography and detection setup was achieved, reaching a transport efficiency of up to 50%. During a four-week long beamtime in 2021, the reaction $^{48}Ca+^{243}Am$ was used again to produce ^{288}Mc in the 3n exit channel. The nuclear reaction products were separated in TASCA and thermalized in a modular Recoil Transfer Chamber (RTC) and transferred into the new miniCOMPACT. Eleven decay chains from ^{288}Mc and ^{284}Nh were registered, adding to the seven decay chains observed in 2020. The complete data set is currently under final evaluation and will provide first information on the interaction strength of Nh and Mc towards SiO_2. The preliminary analysis shows excellent agreement with theoretical predictions [5].

On the way to even more short-lived and chemically reactive elements like Lv and Ts, the extraction time to a detection setup has to be reduced further to the order of a few milliseconds, and reference data from the lighter homologs has to be obtained. A reduction of the transport time can be achieved by using of a buffer gas stopping cell for the thermalization of recoiling ions behind TASCA, in combination with the new miniCOMPACT detector. First test experiments with the existing buffer gas cell from SHIPTRAP showed promising results with short-lived α-decaying Hg, Fr, and At radioisotopes [S. Götz et al., Nucl. Instrum. Meth. B 507, 27 (2021)]. The measured efficiency for transporting chemically reactive Fr radioisotopes into the miniCOMPACT setup confirms the setup to be suitable for the identification of isotopes of volatile as well as non-volatile elements. However, the performance of the existing cell in terms of efficiency and extraction time is insufficient for experiments with superheavy elements beyond Mc due to a low stopping power associated with the limited maximum gas pressure. Based on the published conceptual design of the advanced buffer gas cell "UniCell", first prototypes of the ceramic plates for the UniCell funnel were manufactured at the Institute of Electron Technology in Cracow, Poland. Computer simulations and optimizations using the SIMION package were started, as well as the mechanical design of the buffer gas cell chamber.

To obtain a comprehensive data set on the interaction of the lighter homologs with various surface materials, a new isothermal-chromatography setup has been designed, which will allow interaction studies with various surfaces like quartz, gold, or Teflon in a wide temperature range. This is foreseen for the beamtime 2022.

Chemical theory supporting experimental work

With the aim to interpret results of the experimental work on the reactivity of Fl obtained in previous beamtimes at TASCA, energies, Eads, and other adsorption properties of atoms and oxides of Cn and Fl, as well as of the homologous species of Hg and Pb, on the Au(111) and on fully hydroxylated quartz surfaces were calculated on the basis of 2c-DFT BAND calculations and a periodic slab model. The present results with an improved (dispersion corrected) exchange-correlation functional confirm the earlier predicted sequence in the adsorption of the elemental atoms on the gold surface: Pb >> Hg > Fl > Cn. Oxides of Hg, Cn and Fl should be much more reactive with the gold surface than the corresponding atoms, with ≈200 kJ/mol higher Eads values, except for PbO, where $E_{ads}(Pb) > E_{ads}(PbO)$. A striking difference in the geometry of the adsorbed MO molecules was found between group-12 and group-14 elements. An

analysis of results for adsorption of the M and MO (M = Hg/Cn and Pb/Fl) species on the hydroxylated α-quartz surface shows that Hg, Cn and Fl atoms should not interact with such a surface at room temperature, while Pb should adsorb on it. Oxides of these elements, on the contrary, should strongly adsorb on quartz with E_{ads} of >100 kJ/mol. Thus, for the gas-phase chromatography column with quartz and gold surfaces of the detectors, FlO and CnO should adsorb in the first section on quartz, while Fl and Cn will be transported further and adsorb on the gold surface in the second section, with E_{ads}(Fl) > E_{ads}(Cn). This agrees with experimental results on the elemental species. To study the formation of small molecules of even heavier, expectedly more reactive elements, formation energies and other properties have been calculated via ADF software for compounds such as LvH_2, LvO, TsH, TsOH, OgH and OgOH, etc. and of their lighter homologs. Optimal formation reactions have been suggested.

Quantum chemical method developments

A particular focus of software developments in 2021 was put on enhancing the accuracy and efficiency of (multiconfigurational) relativistic quantum chemical approaches applicable to chemical studies in SHE chemistry. In continuation of our efforts to treat scalar-relativistic effects, spin-orbit coupling and electron correlation effects on an equal footing, we put forward a simplified, yet accurate state-interaction ansatz within the framework of the density matrix renormalization group approach (DMRG) [L. Freitag et al., J. Chem. Theory Comput., 17, 7477 (2021)]. The latter enables faster and more affordable large-scale multiconfigurational calculations with the inclusion of spin-orbit coupling and also makes DMRG-based ab initio excited-state molecular dynamic simulations accessible for future applications. Moreover, to alleviate the computational cost associated with genuine relativistic four-component quantum chemical calculations, we formulated and implemented a new ansatz for two-electron scalar- and spin-orbit picture-change corrections applicable to exact two-component Hamiltonians for relativistic quantum chemistry. First studies on (molecular) compounds of the superheavy elements Cn, Lv and Og showed that the resulting new two-component Hamiltonian models yield molecular properties that are within spectroscopic accuracy (< 1 meV) of the parent four-component data when probing the core (Mössbauer and X-ray ionization spectroscopy) as well as the valence electronic structure (reaction energies and valence excited states).

Actinide target production developments

Superheavy element production relies on optimized long-term stable targets of rare actinide isotopes. Improved methods are needed to produce targets in a thickness range up to >1 mg/cm^2, able to withstand highest beam intensities as provided by ever more powerful accelerators. To this end, a two-prong approach is currently pursued. On the one hand, fundamentals of the most frequently used production technique, molecular plating (MP) are studied and modern electrochemical production processes are evaluated in cooperation with the DECHEMA Research Institute, Frankfurt. On the other hand, microscopic and spectroscopic studies of heavy-ion beam-induced changes in f-element layers are performed together with the Materials Research Department.

Molecular-plated lanthanide targets, both unirradiated ones as well as targets that were previously irradiated with different fluences of 8.6-MeV/u ^{197}Au at the GSI Materials Research department's M-Branch, were analysed with various techniques. Ion-beam analysis performed at the Ion Beam Center at Helmholtz-Zentrum Dresden Rossendorf (HZDR) showed a decrease of the carbon and oxygen content with increasing beam dose. To identify elemental and structural modifications, additional analytical methods, such as infrared spectroscopy (IR), Raman spectroscopy (Raman), X-ray photoelectron spectroscopy (XPS) and grazing incidence diffractometry (GIXD) were applied at the JRC-Karlsruhe in the frame of the RI Open Access scheme ActUsLab; these data are currently under final evaluation. In 2021, a new set of lanthanide targets was irradiated with 5.9-MeV/u ^{48}Ca ions at the TASCA beamline. Some of the samples were produced by the classical MP technique, and others by a novel electrochemical production pathway based on triflate precursors. To facilitate analysis in laboratories not rated for handling radioactive materials, the targets are currently stored to allow decay of beam-induced radionuclides; they will be analysed in 2022.

A multitude of further targets and sources, mostly of actinides, was prepared in the SHE Chemistry section at HIM for studies within our groups, e.g., at TRIGA-TRAP and SHIPTRAP, as well as for use in collaborative work, including Am, Cm, and Cf samples provided for laser spectroscopy studies at the RISIKO laser mass separator. The study of various aspects connected to the exotic low-energy ^{229m}Th isomer is performed in different collaborating groups. For such work, two ^{233}U recoil sources providing the α-decay daughter $^{229(m)}$Th prepared by MP were shipped to KU Leuven, where $^{229(m)}Th^+$ will be extracted by a gas-jet to a laser ionization and spectroscopy setup. Metallic ^{232}Th laser ablation targets were shipped to the University of Granada, where Penning-trap based studies will be performed at the TRAPSENSOR facility, first with ^{232}Th and later with ^{229m}Th. A liquid source of ^{228}Ra placed in a container made from scintillating material was provided to the GSI nuclear spectroscopy department for use at the DESPEC setup. For this,

26.5 kBq ^{228}Ra were separated from about 10 g of ^{nat}Th. For fission studies at the LOHENGRIN mass spectrometer at ILL Grenoble, old targets containing 1 mg of ^{239}Pu were reprocessed. From this material, five new targets with activities of 20-1000 kBq were produced via MP and employed in experiments in 2021. The JRC Geel, Belgium, aims on detailed investigations of undisturbed fission products from spontaneous fission sources like ^{248}Cm and ^{252}Cf, necessitating thin sources of spectroscopic quality placed on extremely thin backings like 10 µg/cm² carbon foils or 24 µg/cm² polyimide foils. The Drop-on-Demand (DoD) printing technique was employed. Parameter studies of the wetting behaviour of different solvents were performed to identify a system that provides thinner deposit layers on polyimide films than the standard water/acid-based system. Ethanol/water mixtures were found to provide superior layers as demonstrated with an ^{243}Am source and verified at JRC Geel. Samples of ^{248}Cm and ^{252}Cf will be produced in 2022. Large-area ^{241}Am calibration sources are needed for the online calibration of a special detector array used in studies of neutron decays at the TRIGA Mainz and the ILL Grenoble. Fourteen such sources with about 6-7 kBq of ^{241}Am each and four with about 25 kBq each were produced by DoD printing on Ti substrates.

Selected publications of 2021

- [1] Khuyagbaatar, J. ; Brand, H. ; Cantemir, R. A. ; et al.: Spontaneous fission instability of the neutron-deficient No and Rf isotopes: The new isotope 249No. Physical review / C 104(3), L031303 (2021), DOI:10.1103/PhysRevC.104.L031303
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- [5] Pershina, V. ; Iliaš, M. ; Yakushev, A._ Reactivity of the Superheavy Element 115, Mc, and Its Lighter Homologue, Bi, with Respect to Gold and Hydroxylated Quartz Surfaces from Periodic Relativistic DFT Calculations: A Comparison with Element 113, Nh. Inorganic chemistry 60(13), 9796 - 9804 (2021), DOI:10.1021/acs.inorgchem.1c01076