4.4 Superheavy elements at GSI and HI Mainz

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In 2022, 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 Fr and Bi nuclei and their progeny, 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.

Highlights in 2022

Synthesis/Nuclear reactions

An intense experimental nuclear reaction study program is carried out by the SHE-Chemistry department within an international collaboration. Analyses of experimental data on nuclear reaction dynamics by detecting fission fragments in the CUBE setup of the Australian National University's (ANU) Heavy Ion Accelerator Facility, Canberra, Australia are ongoing and will soon be published. Also, analyses of experimental data on multi-nucleon transfer reactions performed at the mass separator MARA of University of Jyvaskyla, Finland, were carried out.

Nuclear Structure

At TASCA, nuclear structure studies with the TASCA nominal large area focal plane detector are actively ongoing. In 2022, the experimental program on the study of K isomeric states was successfully continued, focusing on ^{250}No. In this nucleus, an isomeric state with a half-life of about 30 µs, which is longer than its 4-µs SF decaying ground state, is well-known. A direct fission from this isomeric state has not been observed yet, and an upper limit of 0.5 for the fission branch is established.

In the experimental work at TASCA [J. Khuyagbaatar et al., Phys. Rev. C 106, 024309 (2022)], a sensitive search for direct fission from the K isomeric state was performed. No direct fission branch from the isomeric state was identified, and a stricter value of 0.035 for the upper limit was deduced from data obtained in 2018. This shows that the fission from the high-K state can largely be hindered, and the hindrance-strength depends primarily on the properties of an individual nucleus. In addition, a signature for the presence of a higher-lying K isomeric state with a half-life of about 0.7 µs and decaying into the lower-lying one was observed. Based on this observation a second and higher-lying isomeric state has been suggested to exist in ^{250}No.

The intense experimental program studying high-K states in heaviest nuclei [J. Khuyagbaatar et al., Nucl. Phys A 994, 121662 (2020), Phys. Rev. C 103, 064303 (2021)] showed digital electronics to be advantageous for their identification and study. On the other hand, direct fission from a K-isomeric state in the heaviest nuclei is only known for two cases: ^{256}Fm and ^{254}No. These, however, do not represent the most extreme cases of bound many-body quantum systems. Therefore, the fission hindrance factor of high-K states is still poorly understood.

On the theoretical side, an approach to calculate the hindrance strength of the high-K number has been suggested and fission half-lives of various known and predicted high-K states in heavy and superheavy nuclei were calculated [J. Khuyagbaatar, Eur. Phys. J. A 58, 243 (2022)]. The results agreed well with various experimental data on the fission-stability of known K isomeric states. High-K states of nuclei with extremely short ground-state half-lives in the sub-µs range might ensure their survival during transport from production to detection. Therefore, the investigation of the high-K phenomena in the heaviest nuclei at TASCA is continuing.

Meanwhile, an alternative method for the spectroscopy of the heaviest nuclei, which was suggested by the GSI's SHE-Chemistry department in 2020, is still under intensive development. In the 2022 FAIR Phase-0 beamtime, an upgraded version of the Adsorption-based Nuclear Spectroscopy Without Evaporation Residue Signal (ANSWERS-v1) had been used for studies of Ac, Fr, Np and No isotopes. The obtained experimental data again confirm the spectroscopy potential of ANSWERS-v1. The data analysis is ongoing.

The previous experimental data accumulated with the ANSWERS-prototype in 2020, were finalized and are in preparation for publication. The analysis of the experimental data, e.g., the algorithm for trace-analysis, has been significantly improved. In addition, to advance our understanding of the system's response for multi-coincident events involving α-particles, electrons, protons and/or photons, simulations with the GEANT4 toolkit are being developed. The first version in which the ANSWERS-prototype was implemented was introduced in 2022 and its first simulated results are in fine agreement with the experimental responses. These results will be published and be used for the future optimization of ANSWERS.

The analysis of the SHIPTRAP data from the beamtime 2021 proceeded with the emphasis being on investigating low-lying isomeric states in the members of the α-decay chains ^{206}Fr – ^{202}At – ^{198}Bi and ^{204}Fr – ^{200}At – ^{196}Bi. In each of these nuclides two (low-lying) isomeric states were known from investigations by laser spectroscopy [K. M. Lynch et al. Phys. Rev. C 93, 014319 (2016)]. However, the excitation energy of these isomers remained unknown. We have observed both isomers and the ground state in all six nuclides and can directly determine the isomers' energies from the measured mass difference. The investigation of systematic uncertainties in mass measurements with SHIPTRAP has been advanced to support the analysis of the data from the 2021 beamtime and from offline measurements. The latter comprised measurements of decay products from ^{225}Ac and ^{223}Ra sources as well as mass measurements of ^{238}U, ^{242}Pu, and ^{244}Pu. The studies of the systematic uncertainties address mostly effects that are specific for the phase-imaging ion-cyclotron-resonance technique. Results from the 2018 SHIPTRAP campaign were recently published [O. Kaleja et al. Phys. Rev. C 106, 054325 (2022)]. Precise values of the masses of several No, Lr, and Rf isotopes were reported that also allowed improving the masses of 15 additional (super)heavy nuclides up to ^{271}Rg in the framework of the Atomic Mass Evaluation.

Atomic Physics

Laser spectroscopy studies on the heaviest elements continued in two experimental campaigns in April and May 2022. Using ^{40}Ar and ^{48}Ca beams from the UNILAC at GSI, atomic transitions in fermium (Z=100) and nobelium (Z=102) were investigated and the search for an atomic transition in lawrencium (Z=103) was continued. Due to recent advancements of the well-established RADRIS technique [J. Warbinek et al., Atoms 10 (2022) 41], more rare and short-lived nuclides like ^{251}No ($T_{½}$=0.8 s) and ^{246}Fm ($T_{½}$=1.4 s) could be studied. This, in combination with off-line measurements at Johannes Gutenberg University Mainz (JGU) allowed studies on a total of eight fermium isotopes ranging from ^{245}Fm to ^{257}Fm. The data analysis has been completed and a publication is in preparation. Further measurements at JGU comprised the study of long-lived isotopes of californium (Z=98) [F. Weber et al. "Probing the atomic structure of californium by resonance ionization spectroscopy." Atoms 10 (2022) 51] and einsteinium (Z=99) [F. Weber et al. "Atomic-structure investigations of neutral einsteinium by laser resonance ionization." Physical Review Research 4.4 (2022): 043053; S. Nothhelfer et al. "Nuclear structure investigations of $^{253–255}$Es by laser spectroscopy." Phys. Rev. C 105 (2022) L021302] revealing information on their atomic and nuclear structure. This work is a prerequisite for future measurements of short-lived Cf and Es isotopes at GSI.

In addition, an online commissioning experiment of the newly developed JetRIS setup [S. Raeder et al., Nucl. Instrum. Meth. Phys. Res. B, 463 (2020) 272] was performed. This novel apparatus was designed at HI Mainz to improve the spectral resolution for heaviest-element studies. To optimize the laser spectroscopy with improved resolution in the low density and low temperature environment of a supersonic gas jet effusing from the gas stopping cell the influence of the nozzle was studied in detail [D. Münzberg et al., Atoms 10 (2022) 57]. For the on-line experiment, major equipment such as a high-power and high-repetition rate laser system was contributed from external collaboration partners at KU Leuven and from HI Mainz. A laser spectroscopy experiment on ^{254}No was performed with JetRIS, demonstrating the feasibility of the novel approach improving the spectral resolution over that achievable with RADRIS [M. Laatiaoui et al., Nature 538 (2016) 495; S. Raeder et al., Phys. Rev. Lett. 120 (2018) 232503].

A long-term development at JGU and HIM, the novel Laser Resonance Chromatography (LRC) [M. Laatiaoui et al., PRL 125 (2020) 023002] method, aims at laser spectroscopy of superheavy ions, starting with lawrencium. Briefly, optical pumping of ions drifting in dilute helium is exploited to identify optical resonances. Successful excitation of ion levels triggers pumping to metastable states causing an abrupt change in transport properties that can be measured with drift time spectrometers [M. Laatiaoui et al., PRA 102 (2020) 013106]. For the best performance, experimental

conditions must be found to maximize the mobility difference of the ground state and the excited state. This requires accurate theoretical constraints on the parameters in advance, in addition to preparatory off-line experiments. Using the Multi-Reference Configuration Interaction method, we predicted the electronic structure of Lr^+ and Rf^+ as well as the interaction potentials of the Lr^+-He and Rf^+-He systems [H. Ramanantoanina et al., PRA 104 (2021) 022813], [H. Ramanantoanina et al., Atoms 10 (2022) 48]. The interaction potentials are used to obtain the mobility of the ion drifting in helium in its ground state and lowest excited states. In parallel, simulations for ion bunching and drift have been performed to narrow down the working parameters of the LRC setup [E. Romero-Romero et al., Atoms 10 (2022) 87], which is still in the inauguration process, having installed the cryogenic drift tube and the miniature radio frequency buncher and ion guide. Initial results for transfer and bunching of ablated heavy ions work are promising. Optical pumping of Lu^+ ions will be next to establish the LRC method.

Chemical Studies

In the FAIR Phase-0 beamtime 2022, isothermal gas-phase chromatography experiments with alpha-decaying short-lived isotopes of Hg, At, and Po were carried out at TASCA to provide benchmark data for comparative studies with their superheavy homologs (Cn, Lv, Ts). The isolated isotopes were thermalized in a gas-filled volume behind TASCA and were flushed with noble gas streams through an isothermal chromatography column offering gold, quartz, or Teflon surfaces, the temperature of which could be varied in the range of about -70 to +150 °C. The fraction surviving transport through this column was measured in a COMPACT detector setup. All elements were found to interact strongly with the gold surface in the whole temperature range. At and Hg were mainly volatile on SiO_2, but also showed trends in the external chromatogram that clearly deviate from those resulting from Monte-Carlo-simulations performed for an assumed “mobile adsorption” interaction process. These new findings indicate a much more complex adsorption-desorption process, which could possibly be described as an interplay between chemisorption and physisorption.

Flerovium (Fl, element 114) is the heaviest element for which experimental chemical data have been published so far. To elucidate its chemical character and to settle differences in interpretation of experimental data obtained in two sets of experiments carried out by different collaborations, adsorption studies of Fl on silicon oxide and gold surfaces conducted at GSI were published in 2022 [1]. Evidently, two types of interaction of a Fl species with this surface were observed: chemisorption on Au at room temperature, and physisorption on ice. The inhomogeneous nature of the Au surface present in the experiment plays a key role. Two scenarios are able to explain the experimental observation: differences in bond strength of Fl atoms on an inhomogeneous Au surface, with the strength depending on the surface site, or the formation of a Fl compound (e.g. FlO). Both scenarios allow describing the complete experimental data set available on chemical properties of Fl, and they both imply that Fl forms chemical bonds and is hence rather a metallic element than a noble gas.

To allow studies of more short-lived superheavy nuclides, a fast buffer gas stopping cell to connect chemistry detection systems like COMPACT to TASCA is under construction. Ion trajectories simulations were carried out for this UniCell, which is based on the RF ion-funnel technique. Using optimized experimental parameters an extraction efficiency of about 100% resulted. The extraction time is calculated to be in the range of few ms for ions with a mass of 293 u and charge states of +1 and +2. In addition, simulations were also performed on an RF-only ejector-interface. The ejector extraction efficiency was optimized for various gas flow rates and RF frequencies/amplitudes. The transport time through the ejector is calculated to be about 2 ms. In a next step, COMSOL will also be employed to simulate the ion trajectories in the UniCell setup, and the construction will continue.

Chemical Theory Supporting Experimental Work

With the aim to interpret gas-phase experiments on the volatility of Fl [2] and nihonium (Nh, element 113) and its homolog Tl, calculations on the adsorption energy, E_{ads}, of MOH (M = Tl and Nh) on hydroxylated quartz surfaces were completed with the use of the periodic SCM BAND program suite [M. Iliaš and V. Pershina, Inorg. Chem. 61, 15910 (2022)]. Very good agreement is reached between the calculated E_{ads}(TlOH) of 133 kJ/mol on the fully hydroxylated and of 157 kJ/mol on the partially dehydroxylated quartz surfaces on the one hand and experimental adsorption enthalpies, $-\Delta H_{ads}$, of 134/137±5 kJ/mol (at ~300 °C) and 158 ±3 kJ/mol (at ~500 °C), respectively, on the other hand. Taking into account this perfect agreement, we suggest that all the experimental ΔH_{ads} values for Tl should be assigned to the adsorption/desorption of the TlOH molecule. For NhOH, its adsorption properties on various quartz surfaces should be very similar to those of TlOH, with slightly smaller E_{ads} values.

Adsorption properties of group 1 and 2 elements and their compounds including those of elements 119 and 120 on the gold surface were calculated using a periodic BAND suite. According to the results, the elements of both groups should well adsorb on the gold surface, with the weakest adsorption of E119 (E_{ads} of 262 kJ/mol) and E120 (E_{ads} of 185 kJ/mol) caused by the relativistic stabilization of the 8s AO. For group 1 species, the sequence is $E_{ads}(MH) > E_{ads}(M) > E_{ads}(MOH)$.

To assist current gas-phase experiments on the volatility of At, a homolog of tennessine (Ts, element 117), calculations of E_{ads} of At and its compounds on hydroxylated quartz surfaces were performed using the BAND software. It was shown that elemental At should adsorb on the quartz surface very weakly with E_{ads} of -26.3 kJ/mol on geminal and -19.7 kJ/mol on vicinal silanols, while AtOH should adsorb more strongly with E_{ads} of -35.5 kJ/mol on geminal and -41.2 kJ/mol on vicinal silanols.

Selected publications of 2022

- [1] Yakushev, A. ; Lens, L. ; Düllmann, C. E. ; et al.: On the adsorption and reactivity of element 114, flerovium. Frontiers in Chemistry 10, 976635 (2022), DOI:10.3389/fchem.2022.976635
- [2] Pershina, V. ; Ilias, M.: Reactivity of superheavy elements Cn and Fl and of their oxides in comparison with homologous species of Hg and Pb, respectively, towards gold and hydroxylated quartz surfaces. Dalton transactions 51(18), 7321 - 7332 (2022), DOI:10.1039/D2DT00240J
- [3] Khuyagbaatar, J.: Fission-stability of high-K states in superheavy nuclei. The European physical journal / A 58(12), 243 (2022), DOI:10.1140/epja/s10050-022-00896-3
- [4] Warbinek, J. ; Anđelić, B. ; Block, M. ; et al.: Advancing Radiation-Detected Resonance Ionization towards Heavier Elements and More Exotic Nuclides. Atoms 10(2), 41 (2022), DOI:10.3390/atoms10020041
- [5] Kaleja, O. ; Anđelić, B. ; Bezrodnova, O. ; et al.: Direct high-precision mass spectrometry of superheavy elements with SHIPTRAP. Physical review / C 106(5), 054325 (2022), DOI:10.1103/PhysRevC.106.054325
- [6] Münzberg, D. ; Block, M. ; Claessens, A. ; et al.: Resolution Characterizations of JetRIS in Mainz Using 164Dy. Atoms 10(2), 57 (2022), DOI:10.3390/atoms10020057