4.4 SHE departments and HIM SHE section

Department Heads: Prof. Dr. Michael Block, Johannes Gutenberg University Mainz & GSI; Prof. Dr. Christoph E. Düllmann, Johannes Gutenberg University Mainz & GSI
Authors: Brankica Andelic, Ernst Artes, Michael Block, Pierre Chauveau, Premaditya Chhetri, Daniel M. Cox, Christoph E. Düllmann, Dominik Dietzel, Francesca Giacoppo, Michael Götz, Stefan Götz, Raphael Haas, Mustapha Laatiaoui, Oliver Kaleja, Jadambaa Khuyagbaatar, Tom Kieck, Stefan Knecht, Carl-Christian Meyer, Steven Nothhelfer, Valeria Pershina, Sebastian Raeder, Maximilian Rapps, Dennis Renisch, Elisabeth Rickert, Dirk Rudolph, Anton Såmark-Roth, Luis G. Sarmiento, Jessica Warbinek, Alexander Yakushev

In 2020, activities at GSI focused on the UNILAC beamtime within the FAIR Phase-0 program, comprising decay spectroscopy of Fl, chemistry studies of Nh, and laser spectroscopy of Fm and Lr. In addition, the analysis of data obtained in prior beamtimes was continued. At HIM, the advancement of technical and methodological developments, for example for applications in laser spectroscopy and mass spectrometry as well as radionuclide layer production for various applications was most central. In addition, preparations for the beamtime 2021 have been performed.

Synthesis

The search for new elements beyond Og (Z=118) is currently a hot topic in superheavy element research. Attempts to produce the elements 119 and 120 in the ^{50}Ti+^{249}Bk and ^{50}Ti+^{249}Cf reactions were carried out in 2011/2012 at the gas-filled recoil separator TASCA. These reached low cross-cross sensitivities, however, did not result in the discovery of new elements, likely due to aspects of the nuclear reaction mechanism [J. Khuyagbaatar et al., Phys. Rev. C 102, 064602 (2020)]. The reached sensitivity levels are very valuable to enhance the understanding of the nuclear reaction mechanism occurring in beyond-^{48}Ca induced heavy-ion reactions on actinide targets, which have never resulted in the production of SHE to date. While the search for new elements is not currently pursued at GSI, studies of the nuclear reaction mechanism continue, in collaboration with Australian National University in Canberra, Australia. We investigated various deep-inelastic reaction channels in four different reactions potentially suitable for the discovery of element 120 [H.M. Albers et al., Phys. Lett. B 808, 135626 (2020)]. The measured deep-inelastic channels show quasi-fission to be the dominant reaction outcome, however, at different contact timescales. The longest interaction time was found for the ^{50}Ti-induced reaction, i.e., ^{50}Ti+^{249}Cf, which suggests this reaction to be the most promising one in the entrance channel. Therefore, this result confirms that the attempt to synthesize elements 119 and 120 at TASCA in ^{50}Ti-induced reactions was the proper choice [J. Khuyagbaatar et al., Phys. Rev. C 102, 064602 (2020)].

Nuclear structure

Nuclear fission is one of the main issues determining the stability of superheavy nuclei. In the SHE chemistry department the fission process is intensively studied both theoretically [J. Khuyagbaatar, Nucl. Phys. A 1002, 12195 (2020)] as well as experimentally [J. Khuyagbaatar et al., Phys. Rev. Lett. 125, 142504 (2020)]. In a semi-empirical description of all known half-lives of even-even heaviest nuclei [J. Khuyagbaatar, Nucl. Phys. A 1002, 12195 (2020).], it has been shown that fission might be one of the main decay modes for the yet unknown element 120. Accordingly, this decay mode should also be considered in any discovery experiment on element 120. The obtained results also support that a lowering of the outer fission barrier occurs in the superheavy nuclei, which had been inferred previously from the analysis of the electron capture delayed fission. Thus, the new results support the predicted large Electron-Capture Delayed Fission (ECDF) branching in superheavy elements. The ECDF phenomenon is a promising topic for examining the fission properties of superheavy nuclei. The occurrence of this decay mode has been examined in the new isotope ^{244}Md at TASCA [J. Khuyagbaatar et al., Phys. Rev. Lett. 125, 142504 (2020)]. No ECDF

branch was discovered because the new isotope ^{244}Md decays mostly by α-particle emission. At the same time, we have observed a short-lived fission activity, which could not unambiguously be attributed to a specific physical process and deserves further investigation. The work on ^{244}Md was included in the list of the few most important achievements in the fields of particle-, nuclear and accelerator physics [L. Kleinen, Europa bleibt Herrin der Ringe und Quantenoptik erhellt fortan die starke Kraft, Jahresrückblick Teilchen-, Kern- und Beschleunigerphysik 2020, pro-physik.de, 28.12.2020].

In the wake of the discovery of superheavy elements, nuclear spectroscopy experiments aim at providing anchor points at the uppermost end of the nuclear chart for nuclear structure theory, which otherwise had to solely rely on extrapolations. In two runs in 2019 and 2020, such a nuclear spectroscopy experiment was conducted to study α-decay chains stemming from isotopes of flerovium (element Z = 114). One incentive to study flerovium isotopes is that many, but not all, nuclear structure models or model parametrizations favour Z = 114 as the next magic proton number beyond lead, Z = 82. This was studied in an experiment, in which an upgraded TASISpec decay station was placed behind the gas-filled separator TASCA. The fusion-evaporation reactions ^{48}Ca+^{242}Pu and ^{48}Ca+^{244}Pu provided a total of 32 flerovium-candidate decay chains in effectively 18 days of beam time. Two and eleven decay chains were firmly assigned to even-even ^{286}Fl and ^{288}Fl isotopes, respectively. The – admittedly unexpected – observations include (i) an excited 0^+ state at 0.62(4) MeV excitation energy in ^{282}Cn, and (ii) a Q_α = 9.46(1) MeV decay branch, b_α≈2%, from ^{284}Cn into ^{280}Ds [A. Såmark-Roth et al., Phys. Rev. Lett. 126, 032503 (2021)]. Both observations indicate that there is hardly any shell gap at proton number Z = 114 - at least not at neutron numbers N ≈ 172-174. This statement is supported by demanding beyond-mean-field model calculations, which include the necessary triaxial shapes [J.L. Egido and A. Jungclaus, Phys. Rev. Lett. 125, 192504 (2020)]. The existence of the excited 0^+ state in ^{282}Cn requires an understanding of both shape coexistence and shape transitions for the heaviest elements. Second, using the known Q_α = 10.79(4) MeV for the ^{292}Lv -> ^{288}Fl α decay as well as the now precisely measured Q_α = 10.06(1) MeV for ^{288}Fl -> ^{284}Ds, a smooth Q_α sequence across Z = 114 could be established. Hardly any kink is observed at Z = 114, while it is characteristic for any pronounced shell gap. The present results thus reinforce the benchmarking capability of nuclear spectroscopy experiments in the superheavy element regime. Future technical developments on beam intensity, target integrity, and detection efficiency should allow to "wring out tantalizing physics from compound nucleus production data where cross-sections are in the picobarn range" (quote referee report of [A. Såmark-Roth et al., Phys. Rev. Lett. 126,032503 (2021)]).

Nuclear isomers provide valuable information about excited states in nuclei and the interaction or coupling of nucleons staying in different nuclear levels. Of specific interest in well-deformed nuclei are *K*-isomers, arising from breaking up nucleon pairs and exciting them into different nuclear states. In a recent experiment we investigated possible population of *K* isomers in ^{255}Rf produced in the reaction ^{207}Pb(^{50}Ti,2n)^{255}Rf. To detect events from the decay of such isomeric states, correlations of the type Evaporation Residue ER (implantation signal) - CE (conversion electron, possibly in prompt coincidence with a γ event) - α decay / Spontaneous Fission SF (from ^{255}Rf or ^{251}No) were analysed. As one has to consider a possible population of the known $5/2^-$ isomeric state in ^{255}Rf (E* ≈ 135 keV, $T_{1/2}$ = 50 ± 17μs [S. Antalic, F.P. Heßberger, D. Ackermann, S. Heinz, S. Hofmann, B. Kindler, J. Khuyagbaatar, B. Lommel, R. Mann, Eur. Phys. J. A51, 41 (2015)], which essentially decays by CE emission of E < 150 keV, the energy range of CE from decay of possible *K* isomers was restricted to E > 200 keV. Two activities were identified [P. Mosat, F.P. Heßberger, S. Antalic, D. Ackermann, B. Andel, M. Block, S. Hofmann, Z. Kalaninova, B. Kindler, M. Laatiaoui, B. Lommel, A.K. Mistry, J. Piot, M. Vostinar, Phys. Rev. C 101, 034310 (2020).]: a) one with a half-life of $T_{1/2}$ = 38 +12/-7 μs and CE energies essentially below 370 keV, and b) a second one with a half-life of $T_{1/2}$ = 15+6/-4 μs and CE energies essentially above 370 keV. Based on the observation of three correlations of the type ER - CE1 - CE2 - α/SF the events with the longer half-life were attributed to an isomeric state at E* ≈ (1.15-1.45) MeV, the one with the shorter half-life to a state at E* ≈ (0.9-1.2) MeV. For some of the CE also γ events were observed in prompt coincidence, but no line structure was visible due to low statistics. Also, no spin and parity values could be determined. This has to be left for further, more detailed studies.

Atomic physics

The investigation of the heaviest elements by laser spectroscopy at SHIP has been further extended. In the GSI beamtime 2020, the atomic level search in lawrencium Lr was started using the RADRIS method. It was shown that hafnium Hf is a suitable filament material for the efficient neutralization and evaporation of Lr with low background level from surface ionization. A significant part of the range, in which atomic transitions were predicted by atomic theory, was scanned using a two-step laser excitation scheme, but no evidence for a transition was observed, yet. In addition to the Lr level search, for the first time laser spectroscopy of $^{248-250}$Fm was performed by a variant of the RADRIS method. These fermium Fm isotopes are inaccessible in a direct production scheme and were produced via α decay of $^{252-254}$No. A two-step laser excitation scheme was used with the second step populating an autoionizing state. We measured the isotope shift of the 5,112 cm^{-1} transition in $^{248-250}$Fm. However, the hyperfine splitting of this transition in ^{249}Fm was not fully resolved due to limited statistics. This work extended optical spectroscopy in the Fm isotopes to neutron-deficient isotopes below the N = 152 deformed neutron shell closure complementing recent work at the RISIKO mass separator of Mainz university performed in collaboration with the SHE departments at GSI and HIM. There, the long-lived isotope ^{257}Fm was studied by resonance ionization laser spectroscopy in a hot-cavity ion source offline. The same technique was used to study the einsteinium isotopes $^{253-255}$Es. In 2020, an injection-seeded laser was used to measure the complex hyperfine structure splitting of three different transitions in ^{254}Es with increased spectral resolution. The obtained data will provide information of the nuclear moments and the changes in the mean-square charge radii in the Fm and Es isotopes.

For future on-line measurements on the heaviest elements with improved spectral resolution, a new setup for in-gas jet laser spectroscopy was commissioned. Different de Laval nozzles were compared to identify the optimum pressure conditions to form a gas jet with a Mach number of 8. A spectral resolution of about 400 MHz was obtained measuring resonances in the stable isotope ^{174}Yb, a chemical homologue of No. This paves the way for a measurement of the K=8$^-$ isomer in ^{254}No in the 2022 beamtime at GSI.

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

The first chemical study of Nh at TASCA was attempted in 2016. The non-observation of any Nh events in a three-week experiment pointed at losses of Nh atoms on the way to the detection system COMPACT due to a high chemical reactivity of Nh. Since then, several preparatory studies with chemically reactive metals (Tl, Pb, Fr) have been performed in 2018 and 2019, aiming at a faster and more efficient extraction of short-lived and chemically reactive species into the gas chromatography and detection setup, extended with a new detector array (16-element miniCOMPACT), which was directly attached to the recoil transfer chamber. Based on the obtained positive results an advanced 64-element miniCOMPACT detector was built. During a beamtime of 20 days, the reaction ^{48}Ca + ^{243}Am was used to produce ^{288}Mc in the 3n exit channel. Reaction products were separated in TASCA, thermalized in a newly designed recoil transfer chamber (RTC) and transferred into the new advanced miniCOMPACT. Seven decay chains, assigned to ^{288}Mc and ^{284}Nh, were observed. The obtained data are under evaluation and will allow to define the interaction strength of Nh towards SiO_2. The observation of two decay chains originating from ^{288}Mc ($T_{1/2} \approx 170$ ms) is promising for the chemical study of Mc, which is scheduled at GSI for 2021.

Complementing the experiments with the superheavy elements, off-line studies with Pb and Bi as lighter homologs of Fl and Mc were carried out. The short-lived volatile ^{219}Rn was provided from an ^{227}Ac source, which was flushed with flowing gas. The daughters ^{211}Pb and ^{211}Bi were flushed through the RTC into the miniCOMPACT array. Chromatograms were recorded as a function of various parameters like carrier gas type, gas flow rate and pressure, thus characterizing the novel detector array and aiding to optimize the conditions for experiments with superheavy elements. Pb and Bi showed the expected high reactivity towards the silicon dioxide surface of the miniCOMPACT. Furthermore, experiments with oxygen as a reactive gas were carried out. No measurable differences in the distribution were found, suggesting a similar reactivity of the possibly formed lead/bismuth oxides towards the detector surface as the pure elements.

On the way to even more short-lived elements like Lv, the extraction time to transfer the isotopes from the RTC into COMPACT needs to be reduced, as this is significantly longer than the half-lives of even the most long-lived isotopes of all elements beyond Fl. To overcome this limitation, exploratory experiments to study the performance of a buffer gas cell in combination with a COMPACT array were carried out using short-lived α-decaying Hg, Fr, and At radioisotopes. These were produced in ^{40}Ar- and ^{48}Ca-induced nuclear fusion-evaporation reactions, isolated in the recoil separators MARS at Texas A&M University (USA), and TASCA at GSI and thermalized in a gas-stopping cell. From the latter, the nuclear reaction products were extracted into gas-phase chromatographic systems. The efficiency for transporting chemically reactive Fr radioisotopes into the optimized miniCOMPACT gas-chromatography setup was measured and supports that this technique enables the identification of isotopes of volatile as well as non-volatile elements. These studies guide the path towards chemical investigations of superheavy elements beyond flerovium, which are out of reach with currently used setups. However, the stopping power of the presently used gas cell is insufficient for experiments with superheavy elements. To overcome this problem, the design of the advanced "UniCell" has started.

Chemical theory supporting experimental work

In assistance to the coming experiments on the study of reactivity and volatility of Mc and its homolog Bi, calculations of adsorption properties of Bi and Mc on the surfaces of gold and quartz have been performed with the use of the ADF BAND relativistic periodic code. The results have shown that Mc will adsorb on the hydroxylated quartz surface similarly to Nh with an energy of 58 kJ/mol. Adsorption of Bi is much stronger. Both elements will adsorb very strongly on the gold surface with the energy above 200 kJ/mol. With the aim to support gas-phase experiments on the volatility of element 113, Nh, and its homolog Tl, adsorption energy calculations of TlOH and NhOH on the hydroxylated surface of quartz have been performed with the use of the ADF BAND and Quantum Espresso (QE) pseudopotential methods. The preliminary results (-83 kJ/mol with BAND and -69 kJ/mol with QE for TlOH) show that the interaction has a complex nature and that a rearrangement of the surface OH groups occurs. There could also be a formation of H_2O molecules on the surface, from hydrogen atoms and hydroxy groups "ripped" off the surface. This work is ongoing. Further work focuses on the theoretical investigation with molecular ADF on the formation of small molecules of Mc and Ts in COMPACT-type setups. We found that formation of oxides and their reduction with hydrogen is energetically favorable.

Quantum chemical software developments

Quantum chemical studies with a particular focus on molecular properties and reactivities of chemical systems with SHE necessitates treating scalar-relativistic effects, spin-orbit coupling and electron correlation effects on an equal footing. A central aspect of our software developments is therefore to extend the scope of relativistic quantum chemical approaches to SHE in the context of novel electron correlation methods with a particular focus on multiconfigurational approaches. Recently, we presented an implementation of the matrix-product state (MPS) wave function parametrization of the density matrix renormalization group (DMRG) approach within the relativistic quantum chemistry software package DIRAC [S. Knecht, Nachr. Chem. 67, 57 (2019)] which was applied to study electron correlation effects on the valence electronic shell structure of Og [S. Knecht, Nachr. Chem. 67, 57 (2019)].

The DMRG approach provides a polynomially-scaling, efficient means to obtain near-exact full configuration interaction solutions for (very) large active orbital spaces in a systematic and variational manner. Although the DMRG ansatz in an MPS formulation is capable of efficiently treating the static electron correlation problem, ultimately striving for chemical accuracy in the description of chemical reactions and/or spectroscopic properties for SHE requires to take into account dynamical electron correlation. Moreover, orbital relaxation often plays a crucial role for a correct description of chemical properties in ground- and electronically excited states. Hence, work is ongoing describing how the orbital relaxation problem for a relativistic MPS reference wave function can be efficiently tackled in a fully relativistic framework and to address the dynamical electron correlation issue along the lines of non-relativistic developments.

Measurements of fission products from the nuclear reaction between ^{238}U and laser-induced protons at PHELIX

A collaboration between the research departments Plasma Physics and SHE Chemistry succeeded in detecting nuclear reaction products produced by bombarding thin ^{238}U targets with protons generated by short pulses (500 fs) of the high-intensity PHELIX laser (200 J) [P. Boller et al., Sci. Rep. 10, 17183 (2020).]. This process produced, among others, the volatile fission fragments iodine and xenon, which were transported from the target chamber to an activated carbon filter by means of a fast gas-jet transport as it is often used in the chemical study of superheavy elements.

Actinide target production developments

To date, actinide targets for research into superheavy elements are mainly produced using the Molecular Plating (MP) process. Neither the exact chemistry nor the layer modifications induced by heavy-ion irradiation have been fully elucidated so far. For this purpose, MP-produced lanthanide targets were irradiated at different fluences in TASCA and at the M3 branch of the Material Research beamline. Further irradiations with ultra-low-energy particles were carried out at the Offline Deposit Irradiation (ODIn) setup [R. Haas et al., Nucl. Instrum. Meth. A 957, 163366 (2020)] at HIM. The analysis of the irradiated targets with various methods in house and at external partner institutions is ongoing.

To overcome the limitations of classical molecular plating, which is applicable for the production of thin layers with a thickness of at most about 800 mg/cm^2, a novel, triflate-based electrochemical deposition method for target production is being developed. For this, triflate salts of different lanthanides have been synthesized and were activated in the TRIGA reactor. The lanthanide precursors produced in this way were used for extensive experiments series on the electrodeposition from N, N-dimethylformamide (DMF). Layers with thicknesses up to 2000 mg/cm^2 were obtained and were characterized by autoradiography. Circular Tm layers were produced and irradiated with ^{48}Ca beam at TASCA. Characterization is currently ongoing, in collaboration with the Materials Research departments at GSI and at TU Darmstadt. As a next step, this process will be adapted to produce layers in the shape of the TASCA target segment.

Developments for mass measurements of superheavy nuclides

At SHIPTRAP, the vacuum system was further improved by the installation of non-evaporable getter pumps to enable longer measurement times while minimizing losses due to charge exchange and molecule formation. As a consequence, the rate of ^{257}Rf ions available in the SHIPTRAP measurement trap was increased in the 2020 beamtime by almost an order of magnitude compared to 2018 in a preparatory experiment for high-precision mass measurements of Rf and Db isotopes with SHIPTRAP in 2021. Developments for single-ion mass measurements of rare exotic nuclides utilizing a non-destructive electronic detection scheme with the Fourier-transform ion-cyclotron-resonance method were continued. Such methods can be applied in mass measurements of superheavy nuclides with SHIPTRAP and on other exotic nuclides with MATS at FAIR. A novel version of a resonant tank circuit with a quartz resonator serving as the inductor was employed to detect trapped ions in the TRIGA-TRAP Penning trap at room temperature. A sensitivity of some ten ions was achieved and a proof-of-principle mass measurement on ^{206,207}Pb ions was performed with TRIGA-TRAP [S. Lohse, J. Berrocal, S. Böhland, J. van de Laar, M. Block, S. Chenmarev, Ch. E. Düllmann, Sz. Nagy, J. G. Ramírez, and D. Rodríguez, Rev. Sci. Instrum 91, 093202 (2020)]. Further improvements are under way to further increase the sensitivity.

Selected publications of 2020

- [1] Albers, H. M. ; Khuyagbaatar, J. ; Hinde, D. J. ; et al: Zeptosecond contact times for element Z=120 synthesis. Physics letters B 808, 135626 (2020), DOI:10.1016/j.physletb.2020.135626
- [2] Khuyagbaatar, J. ; Albers, H. M. ; Block, M. ; et al: Search for electron-capture delayed fission in the new isotope ^{244}Md. Physical review letters 125, 142504 (2020), DOI:10.1103/PhysRevLett.125.142504
- [3] Khuyagbaatar, J. ; Yakushev, A. ; Düllmann, Ch. E. ; et al: Search for elements 119 and 120. Physical review C 102, 064602 (2020), DOI:10.1103/PhysRevC.102.064602
- [4] Mosat, P. ; Heßberger, F. ; Antalic, S. ; et al: K isomerism in ^{255}Rf and total kinetic energy measurements for spontaneous fission of ^{255,256,258}Rf. Physical review C 101, 034310 (2020), DOI:10.1103/PhysRevC.101.034310