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Superheavy element research – Status report 2016

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The main activity at GSI was the ^{48}Ca UNILAC beamtime, where the experiments were devoted to the continuation of the laser spectroscopy study in No [1] and to the chemical investigation of element 113, which was recently named nihonium (Nh).

In 2015, for the first time optical spectroscopy of nobelium atoms was performed at SHIP, making nobelium the heaviest elements for which such studies were feasible. A strong atomic ground state-transition and several Rydberg states in the nobelium atom were identified by resonance ionization laser spectroscopy [1]. The data showed good agreement with theoretical predictions using relativistic coupled cluster and multi configuration Dirac-Fock approaches. Among the atomic and nuclear properties, a limit for the first ionization potential (IP) of nobelium was obtained. However, a more accurate determination of the IP from the convergence of a Rydberg series was hampered by quenching collisions with buffer gas atoms populating also lower-lying metastable states. In 2016, the laser spectroscopy in nobelium thus focused on the identification of different Rydberg series. To this end, measurements in which the second laser pulse (exciting to a Rydberg state) was delayed compared to the laser for the first excitation step were performed for different pressures in the optical cell. The decay of the RIS signal is indicative of the lifetime of the populated state and allowed us to identify different Rydberg series originating from either the $^1P_1$ state or the metastable $^3D_1$ state. From the convergence of the Rydberg series now the IP of nobelium can be determined with high precision. The data analysis is close to completion and the results will be subject of a forthcoming publication. A rate equation model describing the quenching process was developed and showed good agreement with the data. In addition, the location of the $^3D_1$ state that cannot be excited directly from the ground state was determined indirectly [2]. In the second part of the beamtime, first steps towards laser spectroscopy in the next heavier element, lawrencium, were performed. The stopping and neutralization followed by the evaporation from different filaments was investigated to optimized the conditions for the level search in Lr. In addition, the feasibility of producing ^{255}No by EC decay of ^{255}Lr for laser spectroscopy was demonstrated. This will allow us to extend the measurements of nuclear properties in the nobelium isotope chain.

Besides the laser spectroscopy experiment, the SHE physics department was engaged in several technical developments and upgrades of the setup. The relocation of the

SHIPTRAP setup was completed to fully integrate the new cryogenic stopping cell. The new gas cell operated at 40 K will boost the overall efficiency of SHIPTRAP by up to an order of magnitude and extend the reach for Penning trap mass measurements to heavier elements available with lower yield. In 2016, extensive commissioning experiments with radioactive source (offline) and in parasitic beamtime with ^{254}No were performed. In addition, the recently developed novel phase imaging method (PI-PICR) was further improved by installing new extraction optics. First online mass measurements with upgraded SHIPTRAP system are foreseen for 2018.

The new focal plane detector system for decay spectroscopy at SHIP was characterized in parasitic beamtime by measuring α- and α-γ – decay of ^{253,254}No produced in irradiations of ^{207,208}Pb with ^{48}Ca. The measurements were followed by studies of neutron-deficiency Np isotopes, produced in irradiations of ^{181}Ta with ^{48}Ca where specific emphasis was devoted to the isotopes ^{225,226}Np. The investigation of nuclides in this region near the N=126 shell at the proton drip line can be extended with the new detector system. This system features practically dead time free digital electronics and thus gives access to short-lived nuclides. The data analysis is ongoing. The further analysis of the decay studies of ^{257}Rf and ^{258}Db performed in 2014 resulted in confirmation of two low-lying isomeric states in ^{258}Db, the identification of two short-lived isomeric states in ^{258}Rf, populated by EC decay of ^{258}Db [3], and a low-lying isomer in ^{257}Lr, populated by EC decay of ^{257}Rf [4].

At TASCA, a first attempt on the chemical study of Nh (nihonium, element 113) was performed in 2016. The nuclear fusion reaction ^{48}Ca + ^{243}Am, recently investigated at TASCA in the Mc (moscovium, element 115) decay spectroscopy experiment [5], was selected for the production. The ^{288}Mc recoils were guided through TASCA to an exit window, and were thermalized in a gas flow inside a recoil transfer chamber. The short-lived ^{288}Mc ($T_{1/2}$ = 0.17 s) isotope decayed via alpha-particle emission to ^{284}Nh ($T_{1/2}$ = 0.97 s), the lifetime of which is long enough for the transport to the detection setup. A similar detection setup as in the recent experiments on Fl chemistry was used [6].

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However, to account for the expected higher reactivity of Nh compared to Fl, the first half of the first COMPACT detector array was covered with a SiO_2 surface, while the second half as well as the full second COMPACT array with gold. The latter was cooled to low temperature with a liquid nitrogen cryostat. Fl chemistry experiments at TASCA have demonstrated that one Fl decay chain per week can be observed. Similar cross sections for the production of Fl [7] and Mc [5], as well as comparable beam intensities and target thicknesses led us to expect the observation of about two to three decay chains originating from ^{288}Nh, if the volatility and reactivity of Nh is similar to that of Fl. However, no Nh atoms were detected. The final data analysis is ongoing.

A second chemical system, where studies continued in 2016, is that of carbonyl compounds of transition metal complexes, with $Sg(CO)_6$ having been first synthesized in 2013 [8]. Current techniques include the Sg synthesis, its isolation in a recoil separation, followed by chemical synthesis of the compound behind the separator [8]. A next step included an experiment designed to measure the thermal stability of the compounds in the spirit of [9], which was performed under the lead of the heavy element group from Paul Scherrer Institute (PSI), Villingen, Switzerland, at the RIKEN Nishina Center, Wako, Japan. The focus of the work of our group was on further developments to extend studies of carbonyl compounds of the heaviest elements to those beyond Sg. Fusion products from the asymmetric nuclear fusion reactions, as needed for the carbonyl studies with Sg, Bh, and Hs, have a relatively large angular and energy spread, thus the transmission efficiency through an on-line recoil separator is relatively low. In case of TASCA or GARIS (which was used in [8]), the efficiencies are in order of 13% for Sg [10]. Thus, the overall efficiency of the synthesis of carbonyl complexes in combination with physical preseparation is rather low. For future experiments, the possibilities for chemical investigation of the metal carbonyl complexes of SHE without using a preseparation stage are currently being explored, with the goal to avoid the corresponding losses of close to 90%. First experiments performed at the Tandem accelerator at JAEA Tokai, Japan, suggested the successful the synthesis of Os and W carbonyl complexes also without a preseparator to be feasible, if the thermalization of the evaporation residues is spatially decoupled from the chemical synthesis. The latter is required to occur in a beam-free environment [11]. Further experiments were performed at the research reactor TRIGA Mainz [12].

With the aim to support gas-phase experiments on study of stability and volatility of carbonyls of the heaviest elements, calculations of the electronic structure and properties of group-6 $M(CO)_6$ [13] and group-8 $M(CO)_5$ [14] have been performed using the most advanced relativistic quantum-chemical methods (ADF BAND, X2c-DFT, DIRAC). The results have shown that in contrast to earlier published works the carbonyls of Sg and Hs should be less stable than those of the lighter 5d-homologs. This finding is valuable for fixing the right conditions in measurements of the first bond dissociation energies of these complexes. In addition, using results of these calculations, volatilities of goup-8 carbonyls as adsorption energies on inert surfaces have been predicted via a model of mobile adsorption. It was shown that $Hs(CO)_5$ should slightly more strongly adsorb on neutral surfaces than $Os(CO)_5$.

To render assistance to gas-phase experiments on study of reactivity of elements 112 through 114 with various surfaces, calculations of the adsorption energies of these elements and their lighter homologs on a hydroxylated quartz surface have been performed using a periodic ADF BAND code [15, 16]. Such periodic calculations of adsorption energies have been performed for the first time for superheavy element systems. The results have shown that Cn should be indeed the most volatile element out of those under consideration. Also, Fl should not interact with quartz at room temperature. Element 113, Nh, on the contrary should strongly interact with quartz [16]. Such a different adsorption behavior allows for a good separation between all these elements using a combination of quartz and gold surfaces.

A further activity of the SHE Chemistry division concerned the development of a new detector system for ALpha-BEta-Gamma (ALBEGA) multicoincidence spectroscopy for chemically separated samples [17]. Efforts in 2016 were mainly dedicated to the study and development of the new alpha/beta detector. The new version will be characterized by a more densely packed configuration and a thinner dead-layer on the side in contact with the gas flux. The production of the device was performed at the ITE, Warsaw (Poland), for which an ad hoc technological development for its assembling was required. The new alpha/beta detector will not only provide a higher energy resolution and efficiency, but will also feature increased mechanical stability to sustain the pressure difference inside and outside of the gas channel.

Presently, one of the hot topics in the superheavy element research is the synthesis of elements beyond Og (Z=118). The lack of sufficient amounts of heavier actinides prevents a continuation beyond Og with ^{48}Ca-induced reactions. An obvious path is to continue with fusion-evaporation reactions, but with projectiles heavier than ^{48}Ca. Several experiments have already been performed to synthesize Z=119 and 120 by using the reactions ^{50}Ti+^{249}Bk (TASCA, GSI), ^{64}Ni+^{238}U (SHIP, GSI), ^{58}Fe+^{244}Pu (DGFRS, FLNR, JINR), ^{54}Cr+^{248}Cm (SHIP, GSI), and ^{50}Ti+^{249}Cf (TASCA, GSI). In total, about one year of accelerator beam time has been spent for these search experiments. However, none of them led to the discovery of a new element, suggesting that cross sections are significantly lower than for ^{48}Ca-induced reactions. To get a better guidance for future search experiments, a better understanding of the fusion reaction is needed. Therefore, to understand the reaction mechanism better, an intensive experimental campaign involving various heavy

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projectiles and actinide targets was carried out by a collaboration of scientists from GSI, HIM Mainz, Johannes Gutenberg University Mainz, and the Australian National University (ANU), Canberra, Australia at the ANU's Heavy Ion Accelerator Facility. Suitable actinide targets like ^{244}Pu, ^{248}Cm, and ^{249}Cf were produced at the Institute of Nuclear Chemistry at the Johannes Gutenberg University Mainz, and were irradiated with a variety of beams between ^{34}S and ^{64}Ni at energies around the Coulomb barriers. The mass and angular distribution of fission fragments originating from the nuclear reactions were measured, in many reactions for the first time. The data analysis is ongoing and preliminary results already show a difference in dynamics of reactions involving different projectiles.

Further studies of the fission mechanism were performed, e.g., a study of fission induced by ^{18}O+^{249}Cf multi-nucleon transfer reactions at 8 MeV/A, in which the SHE Chemistry department participated. The experiment was performed at the Tandem accelerator at JAEA Tokai (Japan). Such reactions, acting as surrogate of n-induced fission reactions, allow populating in a single experiment several isotopes at low angular momentum and at low excitation energy. The excitation energy of the fissioning system can be determined by the kinematical reconstruction of the binary process, by measuring the mass and kinetic energy of light ejectile of the transfer reaction. In particular, this experiment was performed with the aim to extend the current systematics of fission fragment mass distribution nearby the region of transition from asymmetric to symmetric fission.

A further activity, which led to widespread recognition, was the contribution of GSI, HIM, and Johannes Gutenberg Mainz scientists and technicians to the direct detection of the exotic low-lying nuclear isomer in ^{229}Th [18]. For this, ^{233}U targets, which yield the ^{229m}Th after alpha decay of ^{233}U, as well as ^{234}U targets (for control experiments serving to exclude an origin other than that of ^{229m}Th decay to the ground state as a source for the observed signal) were produced. They were used in these joint experiments, which were led by the group of P. Thirolf at the Ludwigs-Maximilians-University Munich, Germany.

Some further activities, also including contributions of the SHE Chemistry department to the upgrade of the UNILAC Accelerator, are detailed in individual contributions to this GSI Scientific Report 2016 (J. Konki et al., A. Di Nitto et al., S. Götz et al., M. Götz et al., V. Pershina et al., as well as P. Scharrer et al.)

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The new isotopes ^{240}Es and ^{236}Bk

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Two new neutron-deficient nuclei ^{240}Es and ^{236}Bk were synthesised in an experiment carried out at the Accelerator Laboratory of the Department of Physics (JYFL), University of Jyväskylä, Finland [1]. The hitherto unknown isotopes were identified by their radioactive decay chains starting from ^{240}Es produced in the fusion-evaporation reaction ^{209}Bi(^{34}S,3n)^{240}Es.

Evaporation residues (ER) recoiling out of the target were separated from the primary beam and transfer reaction products by the gas-filled recoil separator RITU [2]. The ERs passed through a Multi-Wire Proportional Counter (MWPC) and were implanted in the two adjacent double-sided silicon detectors (DSSDs) of the GREAT focal plane spectrometer [3] where their subsequent decays were measured.

The energies of all signals from the detectors were time stamped and recorded using the triggerless Total Data Readout (TDR) data-acquisition system [4]. The spatial and temporal correlations in the data between the detectors were analysed using the GRAIN software package [5].

The results of this experiment were reported in Physics Letters B [1] in detail. Two groups of α particles with energies E_α=8.19(3) MeV and 8.09(3) MeV were assigned to ^{240}Es from correlated ER-α events. The new isotope ^{236}Bk was identified from its electron-capture delayed fission (ECDF) branch. Half-lives of 6(2) s and 22^{+13}_{-6} s were obtained for ^{240}Es and ^{236}Bk, respectively. The ECDF probabilities (P_{ECDF}) of ^{240}Es and ^{236}Bk were determined from correlated ER-fission and ER-α-fission events to be 0.16(6) and 0.04(2), respectively. The proposed decay schemes for the new ^{240}Es and ^{236}Bk isotopes are shown in Fig. 1.

The ECDF probabilities of the heavier odd-odd isotopes $^{242-248}$Es and ^{238,240}Bk have been measured previously (see e.g. [6] and the references therein). The new data from this work extend the systematics of the Es and Bk isotopes and show a continuation of the exponential increase of P_{ECDF} as a function of the Q-value of the EC decay (Q_{EC}) and the spontaneous fission barrier (B_{sf}) in more neutron-deficient isotopes. No deviations from this trend

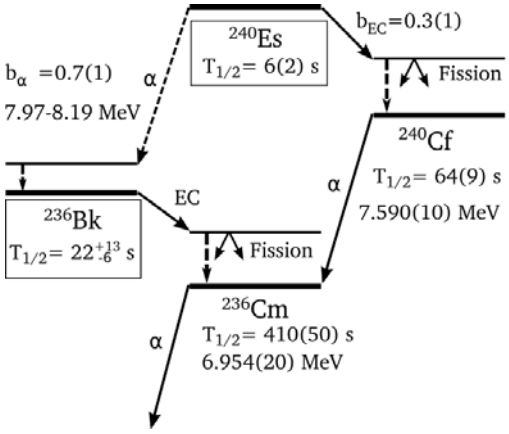

Figure 1: The proposed decay scheme of the new isotopes ^{240}Es and ^{236}Bk. The measured values for ^{240}Es and ^{236}Bk are from this work.

are observed. We note that P_{ECDF}-values for the new isotopes ^{240}Es and ^{236}Bk are the highest ones so far for Es and Bk isotopes, respectively.

The simple dependence of P_{ECDF} on $Q_{\mathrm{EC}} - B_{\mathrm{sf}}$ is not yet fully understood. There is a relative difference between Es and Bk isotopes in P_{ECDF} that could be attributed to the shape of the total fission barrier. In more neutron-deficient isotopes the P_{ECDF} is expected to approach saturation and more experimental data are needed there to shed a light on this complex decay process.

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Hexacarbonyls of Mo, W, and Sg: Electronic Structure and Bonding

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The class of carbonyl compounds has recently been enriched by a new species, a hexacarbonyl of a superheavy element with Z=106, $Sg(CO)_6$ and its volatility has been studied both experimentally [1] and theoretically [2]. Now, experiments are under way to measure the First Sg-CO Bond Dissociation Energy (FBDE) [3]. Earlier predictions based on RECP calculations have indicated that the Sg-CO bond should be stronger than the W-CO one due to relativistic effects on the 6d AOs [4]. With an aim to prove these earlier predictions on a higher level of theory and to have an own set of data, in this work, we have calculated various properties of the group-6 hexa- and pentacarbonyls and have newly determined FBDEs. The following decomposition reaction was considered

$$M(CO)_6(O_h) \rightarrow M(CO)_5(C_{4v}) + CO.$$

The calculations were performed using a variety of nonrelativistic (NR), relativistic scalar (SR) and spin-orbit (SO) methods, such as ZORA-ADF, X2c+AMFI-CCSD(T) and Dirac-Coulomb-DFT one. The uncontracted Dyall vdz basis sets in the DIRAC CCSD(T) calculations were utilized. The present results for the FBDEs in comparison with those of the work [4] and experimental data for $Mo(CO)_6$ and $W(CO)_6$ [5] are given in Table 1.

Table 1. First M-CO Bond Dissociation Energies at various levels of theory (in kJ/mol)

Method	$Mo(CO)_6$	$W(CO)_6$	$Sg(CO)_6$
ADF (NR)	155.07	218.47	223.81
ADF (SR)	163.63	191.19	181.09
ADF (SO)	163.49	190.73	180.22
DC-DFT	163.20	189.83	177.42
X2c+CCSD(T)	158.19	181.45	176.22
RECP-CCSD[a]	170.71	197.90	204.59
RECP-CCSD(T)[a]	182.00	207.94	212.13
ZPT[b]	-5.17	-5.43	-5.59
Exp.[c]	167.4±8	192.5±8	-

[a]Ref. [4]; [b]Zero point and thermal contribution; [c]Ref. [5].

The main difference between the present relativistic ADF, ReSpect and DIRAC calculations on the one hand and the RECP ones [4] on the other hand is an opposite trend in the vibrational frequencies of the M-CO bond and FBDE from $W(CO)_6$ to $Sg(CO)_6$: all the former show a decrease in this direction, while the latter an increase (Fig. 1).

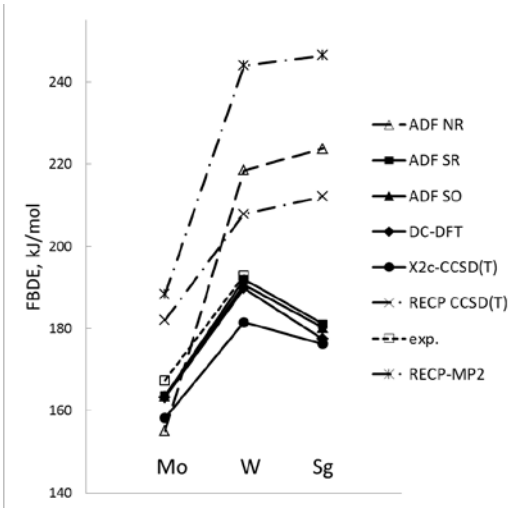

Figure 1. First Bond Dissociation Energies of $M(CO)_6$, where M = Mo, W and Sg, calculated using various methods in comparison with experimental data for the Mo and W carbonyls (open squares).

To find a reason for such a difference, a bond analysis of $M(CO)_6$ (M = Mo, W, and Sg) was performed using ADF Hirshfeld effective charges, q_M, Mulliken MO analysis based on the M and CO fragments and the DIRAC projection analysis. The obtained smaller relativistic q_M(Sg) than q_M(W) means that the electron density is not shifted so much in $Sg(CO)_6$ from Sg to CO as in $W(CO)_6$, so that the Sg-CO electrostatic interaction is smaller than the W-CO one, meaning that bonding is weaker in $Sg(CO)_6$. Non-relativistically, it is just the other way around: the largest q_M on Sg is the source of its largest ionic bonding with CO in the row of homologs. Thus, in difference to the earlier predictions [4], the decomposition of $Sg(CO)_6$ should occur at lower temperatures than those of $W(CO)_6$ [6], with a reversal of the trend from Sg to W.

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Identification of reaction products in ^{50}Ti + ^{249}Cf reactions at TASCA *

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During the last decades heavy ion induced reactions were applied to explore the chart of nuclei up to the superheavy elements (SHE), which has resulted in the discovery of the SHE up to Og ($Z = 118$) in complete fusion reactions [1]. Alternatively, recent model calculations suggest the possibility to produce exotic nuclei including SHE in non-fusion channels of heavy ion induced reactions [2]. Pioneering studies on the possible production of such exotic nuclei in non-fusion reactions were performed in the late 1970s by applying chemical separation techniques [3], which are suitable for longer-lived nuclei ($\gtrsim$1 h). Many properties of the multi-nucleon transfer reactions have been established, but still detailed information on the reaction mechanism/kinematics is missing [5, 6, 4].

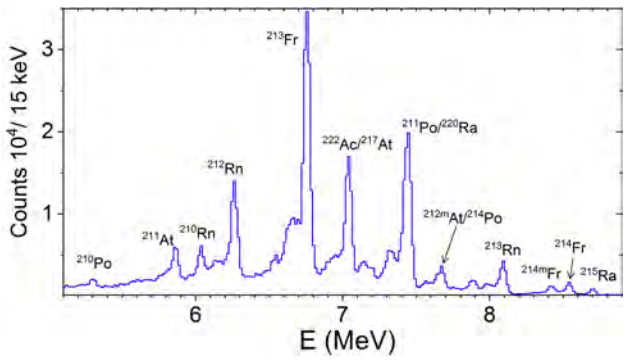

Figure 1: Energy spectrum measured with the focal plane detector during beam-off periods for the ^{50}Ti + ^{249}Cf reaction. Isotopic identification of some lines is given.

Recently, at the velocity filter SHIP new short-lived (down to 10^{-6} h) neutron-deficient nuclei of heavy elements have been synthesized in the ^{48}Ca + ^{248}Cm reaction [7]. These and other results at SHIP (see [8]), benefiting of the kinematic separation, demonstrate the relevance of the forward angle measurements for the investigation of the reaction dynamics.

At the gas-filled recoil separator TASCA, non-fusion products of the ^{50}Ti $+^{249}$ Cf reaction have been investigated. They were produced during the experiment for searching the SHE with $Z = 120$ [9]. The magnetic settings of TASCA were tuned to collect the products of fusion-evaporation reactions, but even under these conditions some amount of non-fusion products were passing through TASCA and were implanted into the focal plane detector. Here their subsequent radioactive decays were measured. A typical energy spectrum containing lines from the α decay of the implanted nuclei is shown in Figure 1. By exploiting the α decay properties, the identification of nuclei was performed employing a position and time correlation analyses between implantation and/or α-like events. In total, 57 isotopes with $Z = 83 - 90$ were identified.

Experimental details and the final analysis will be given in a forthcoming publication [10].

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Speeding up gas-phase chemistry to access elements beyond Fl

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The electronic structure of the heaviest elements is strongly influenced by relativistic effects, which may lead to chemical properties that deviate from those expected based on an extrapolation of trends present in the periodic table [1]. Chemical properties have been reproducibly studied for all elements up to Hs (Z=108) as well as for Cn (Z=112). Current research activities in the field focus on the elements Nh (Z=113) and Fl (Z=114) [2]. Due to the low production rates and short half-lifes, $T_{1/2}$, only single atoms are available in chemical experiments. Nevertheless, the required sensitivity can be achieved, best by combining chemical setups with electromagnetic preseparators [3]. Gas phase chemical methods have proven most successful for the heaviest elements, as they give access to isotopes with half-lifes of the order of at least about one second [2]. The successful gas-chromatography studies of Fl demonstrate the potential of the combination of the chromatography detector array COMPACT with the gas-filled separator TASCA at an one-atom-at-time level [4]. The most time-consuming step in the experiments performed with the current TASCA-COMPACT setup as it was used for the Fl experiments is the thermalization of the fusion-evaporation reaction products in the Recoil Transfer Chamber (RTC) [3] and their transport to the connected COMPACT detector array. Besides Cn and Fl, also Nh is in reach with this technique. The extraction time is, however, significantly longer than the half-lifes of the most long-lived isotopes of all elements wit $Z > 114$. For Mc (Z=115), for example, the most long-lived currently known isotope is ^{288}Mc with $T_{1/2}$ = 170 $\binom{+40}{-30}$ ms [5]. To overcome this limitation, exploratory experiments on the coupling of COMPACT to an existing gas-catcher operating with electric fields [6] were carried out.

To get access to shorter-lived isotopes, it is crucial to speed up the transport time, while maintaining a high efficiency of the setup [5]. To this end, a faster and more effective transport technique is currently being developed. For future experiments with Mc and beyond, the current RTC will be replaced by a gas-catcher, which uses electric fields to extract the ions [6, 7, 8, 9].

In 2016, first off-line measurements were performed with ^{223}Ra ($T_{1/2}$ = 11.43 d) and ^{225}Ac ($T_{1/2}$ = 10 d) recoil ion sources. The used sources were placed in axial symmetry in the center of the direct current electrodes-system (DC-cage), which is located within the gas-catcher. The DC-cage contains the stopping volume for the ions inside the gas-catcher. The system consists of 5 cylindrical electrodes with an outer diameter of 180 mm and an inner diameter of 160 mm. From the source the recoil ions ^{219}Rn$^+$ ($T_{1/2}$ = 3.96 s) or ^{217}At$^+$ ($T_{1/2}$ = 32.34 ms) are guided by the electric fields through the segmented DC electrode towards a funnel structure. This funnel guides the ions by applied DC and AC (RF: 140 V, gradient 7 V cm^{-1}) fields to the exit hole (5 mm diameter). After exiting, the ions are neutralized by collisions with the walls of a Teflon-tube. This neutralization section facilitates gas-chromatography studies of the element of interest in elemental form. After the neutralization zone the atoms are directed by a gas flow into the subsequent COMPACT-detector array [4]. The gas-catcher-COMPACT-setup was flushed with helium gas and kept at a pressure of 50 to 100 mbar. This ensured a viscous flow and allowed performing gas phase chromatography studies in COMPACT. For ^{219}Rn, only decay in-flight was observed as COMPACT was operated at ambient temperature. Due to the high adsorption tendency of ^{221}Fr no atoms of this element were observed in COMPACT in a first test, ^{217}At, however, was observed in the first COMPACT-detector. We studied the efficiency for transporting ^{217}At from the source to the COMPACT detector as well as the transport time. For the efficiency quantification, the measured rate in COMPACT was compared with the effective source strength, which was determined in a separate measurement of the rate at which ^{217}At$^+$ ions are recoiling from the source. To measure the transport time, the potential, at which the source was kept, was cyclically switched rapidly from negative (to avoid release of ^{217}At$^+$ ions) to positive. This was used as the start of a time-of-flight measurement. The stop signal was given by measured decay in COMPACT. Transport times well below 100 ms were measured, which would be fast enough for applications to, e.g., Mc.

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Radiochemical investigation of the kinematics of multi-nucleon transfer reactions

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Renewed interest in multi-nucleon transfer reactions as a promising tool for the production of neutron-rich transactinide isotopes [1] has motivated us to perform a ^{48}Ca+^{248}Cm bombardment at an incident energy 10% above the Coulomb barrier to study emission-angle resolved kinetic energies of isotopes of Bk through Fm [2]. The isotopes of interest were implanted into a stack of Ni foils mounted behind the target. They were isolated off-line using radiochemical separations and detected by alpha spectroscopy. This way, long-lived isotopes with mass numbers between 246 and 256 were accessible. Angular distributions and kinetic energies corresponding to recoil ranges in the Ni foils were evaluated and weighted mean values in the center of mass system were used to construct deflection functions, total kinetic energy TKE vs. Θ_{cm}. Fig. 1 shows the location of the centroid of the Fm activities in the TKE vs. Θ_{cm} plane corresponding to an average total kinetic energy loss TKEL = 70 $\pm$ 10.5 MeV at Θ_{cm} close to the grazing angle. Taking into account the ground state Q value for the most probable primary fragment results in an excitation energy corresponding within the uncertainty in the measured TKEL to the missing mass, i.e. the mass difference between the most probable secondary mass number and the most probable primary mass number calculated with Volkov's generalized Qgg systematics [3] (minimum potential energy corrected for the breaking of nucleon pairs in the multi-nucleon transfer process) indicating that the number of evaporated neutrons is 1.3 on the average. For products closer to the target, e.g. for Cf, the values of TKE spread much wider than for Fm, see Fig. 2. Here, TKEL values of ≈9, ≈34, and ≈54 MeV corresponding to the evaporation of 0, 1, and 3 neutrons are observed in the corresponding range bins.

Thus, values of TKE reach from quasi-elastic to completely damped values. For multi-nucleon transfer products (Fm), the distributions are peaked close to the grazing angle with laboratory kinetic energies close to 80 MeV. These are benchmarks for the design of a large acceptance separator for the separation and detection of short-lived neutron-rich transactinide isotopes.

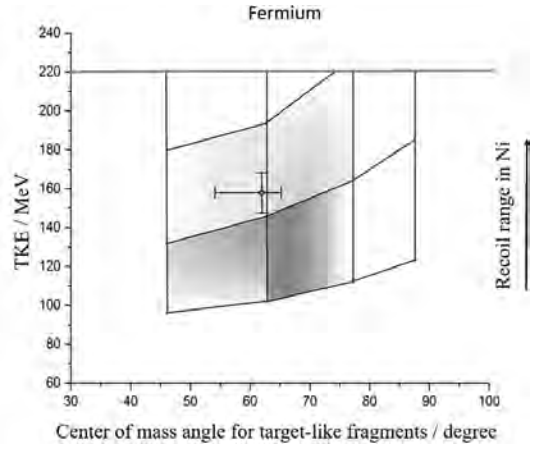

Figure 1: Centroid of the Fm distribution with standard deviations. The shades are meant to indicate the approximate width of the distribution.

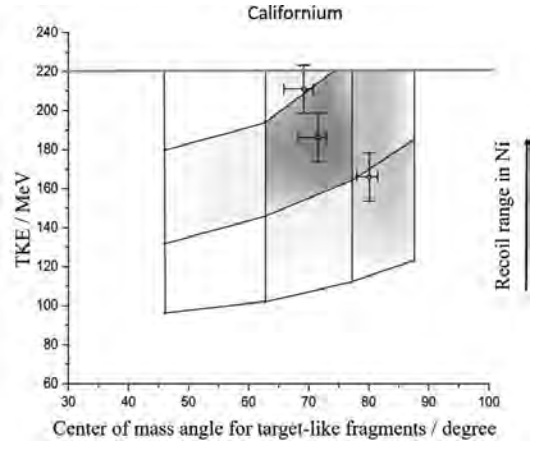

Figure 2: Same as Fig. 1 but for the Cf isotopes.

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Status of the pulsed gas stripper for the UNILAC

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The GSI UNILAC will be used as part of the injector chain for FAIR [1]. In order to meet the high demands in terms of delivered beam current and quality an extensive upgrade program of the UNILAC is ongoing [2]. In this process a new setup for the gas stripper at 1.4 MeV/u was developed [3] and has been first employed in 2014. A pulsed gas injection is used instead of the continuous gas-jet of the previously existing stripper setup. With this setup, using H_2-gas, and together with additional improvements, significantly increased uranium beam intensities were achieved behind the gas stripper [4].

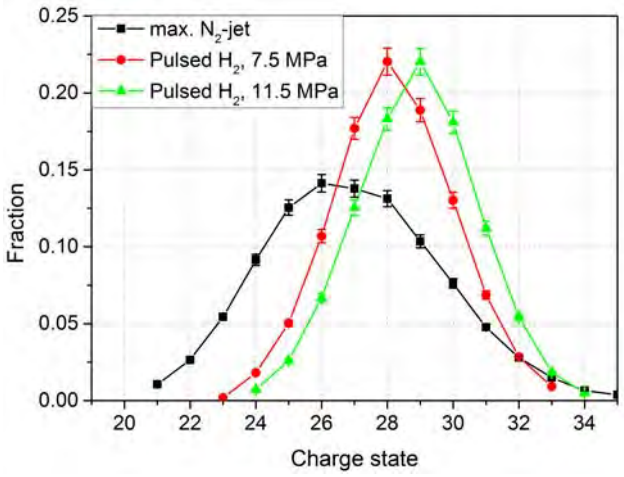

Figure 1: Comparison of the charge state distributions of ^{238}U ions after passing through the charge stripper applying different gas targets and back-pressures on the gas inlet.

Recent measurements

In 2016, additional measurement series were conducted over five days of machine beam time. The main topic was to obtain complete data sets for stripping of uranium and titanium beams, complementing measurements conducted in 2015, and additional measurements of the beam properties behind the gas stripper. For high-current measurements the VARIS-type ion source [5] was used and optimized for high-current operation. With the pulsed gas stripper, using H_2-gas, the target thickness can be adjusted to shift the maximum of the charge state distribution of uranium either to charge state 28 or 29 (see Fig. 1).

Measurements of charge state distributions of uranium and titanium beams after passing through different gases at 0.12 MeV/u and 0.74 MeV/u beam energy were conducted. To enable measurements at lower beam energies, the DTL tanks of the HSI were turned off separately. The corresponding results for 0.74 MeV/u are presented in [6]. At 0.12 MeV/u beam energy, the major part of the charge state distribution could not be measured due to limitations of the magnetic field of the dipole magnet in the charge separation system.

Additionally, the optimal target thickness for operation with the titanium beam was determined to reach maximum yield into a desired charge state. Due to limitations of the differential pumping system, this target thickness is currently not viable in long-pulse operation mode (5 ms pulse length, 50 Hz repetition rate). Alternatively, N_2-gas can be used with the pulsed gas stripper to match the performance of the previously existing N_2-jet stripper setup.

Long-term test

The pulsed gas stripper setup has been used continuously for three weeks during the SIS18 beamtime as a long-term test. During this time, the UNILAC delivered high-intensity U^{28+}-ion beams. A pulsed H_2-gas target with a thickness of about 9 μg/cm^2 was used.

After about two weeks, an increased gas pressure in the adjacent accelerator structures was noticed, which caused the closure of the adjacent gate valves. This was found to be due to a leaky gas valve, which had been in use as the main gas inlet for about 15 days in the course of several beam times and offline measurements in 2015-2016. After switching to the second gas valve, which had significantly less duty time in the past, the beam time continued without any further incidents due to the gas stripper. Additional systematic long-term measurements with the pulsed gas valves are in preparation at a dedicated test stand to identify the cause of this outage.

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Platform development for laser accelerated particle induced nuclear reaction studies utilizing RC methods*

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Introduction

A team of GSI and LLNL researchers successfully completed a platform development experiment to enable future nuclear science experiments at short-pulsed laser facilities. The experiment is a joint effort between Plasma Physics group led by V. Bagnoud and radiochemists from the Super Heavy Element research group at GSI led by A. Yakushev. This first experiment demonstrated an efficient collection of isotopes produced in nuclear reactions with laser driven MeV proton beams. The proposed experiment was awarded the requested run-time of 20 shifts and 40 shots have been used to demonstrate the isotope collection efficiency and reproducibility.

Experiment

The experiment utilizes laser accelerated proton beams (5 – 20 MeV) in combination with radiochemistry based isotope analysis to study nuclear reactions. In particular, the ^{63}Cu(p,n)^{63}Zn reaction was used to measure the proton activated radioactive ^{63}Zn via its β^+ decay and subsequent 511 keV gamma emission identified by its $T_{1/2}$ = 38 sec half-life.

The proton beams were produced through the TNSA (Target Normal Sheath Acceleration) mechanism utilizing GSI's PHELIX laser facility. Laser pulses at 90 J and 500 fs impinging on a thin gold target (the observation of laser accelerated protons has first been reported from experiments at LLNL laser facilities [1,2]. After various tests to identify the spatial, time and energy distribution the accelerated protons were used to activate thin ^{63}Cu-foils and to measure the level of activation, which was found to be consistent with known milli-barn cross sections for 5 – 20 MeV protons. Due to the divergence of the accelerated protons over about 30 mm, the spot size at the interaction with the target foils is around 10 mm. The spot size of the accelerated protons at the exit of the conversion foil is less than 1 mm due to the small source emittance.

After passing through a thin capton foil as debris shield and a thin Ti-foil as window in the gas filled (in flow mode) target cell inside the main target chamber, the protons interacted with ^{63}Cu target foils (1-5 stacked foils) inside the target cell. The produced ^{63}Zn recoil isotopes were stopped in a He/aerosol gas mixture at about 1 bar gas pressure, and transported to a filter through a thin tube. The decay of the ^{63}Zn was identified by measuring the 511 keV annihilation gammas following the β^+ decay with a half-life of 38 min (Fig. 1). The reaction ^{63}Cu(p,n)^{63}Zn and gas transport wsas verified by comparing shots with and without ^{63}Cu foils and with and without carrier gas. The length of the transport line was varied from a 20 to 1 m distance resulting in a transport efficiency varying between 10 to 60%.

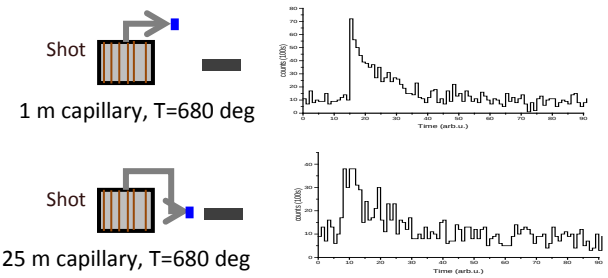

Figure 1: 511 keV gamma emission following the β^+-decay of ^{63}Zn after gas collection utilizing a 1 m (top) and 25 m (bottom) capillary transport line; the decay of the 511 keV gamma line confirms the 38 sec half-life of the ^{63}Zn isotope.

The presented experiment at PHELIX with laser accelerated MeV protons and a pulse duration of around 1 ps opens up new domains of studies through the detection of short lived isomers. The experiments provide critical experimental input for future experiments at FAIR (Facility for Antiproton and Ion Research), but also at laser facilities as ELI (Extreme Light Infrastructure) [3,4]. The collaborative experimental research is part of an agreement between LLNL (DOE) and the GSI (BMBF).

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