# Activities in superheavy element research at SHIP and TASCA

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In the year 2017, efforts of the Superheavy Element Chemistry department focused on analysis of data obtained in the past UNILAC beamtime period. Major modifications to the experimental setups were performed: i) an upgrade of the cave X8 (TASCA) to install improved shielding, ii) the installation of a new, more flexible TASCA detector chamber, iii) a relocation of the TASCA control room in anticipation of major refurbishment of the SE building, which currently houses the control room, and iv) the design and implementation of a next generation TASCA Control System. Thus, best use was made of the extended shutdown period at GSI.

Selected experiments were performed at other facilities, including the Cyclotron Institute at Texas A&M University, College Station, USA.

'For the superheavy element physics department the activities concentrated on the analysis of data obtained in the past UNILAC beamtime period and preparations of the different experimental setups for the planned experiments in the beamtime period 2018 at GSI.

In parallel, several technical developments were advanced, for example, the completion of the decay spectroscopy system COMPASS, the construction of the single-ion mass measurement setup SHIPTRAP-2, and the design of a novel setup for gas-jet laser spectroscopy.

The specific activities are discussed briefly in the following sections.

## *Nuclear reaction studies*

The drastic reduction of fusion-evaporation cross sections for reactions that form the heaviest elements cannot be understood without knowledge of the competing quasifission (QF) process, which hinders the fusion. Generally, this competition is studied by measuring fission fragments originating from QF and fusion-fission (FF). However, quantitative descriptions of experimental results become complex as many degrees of freedoms are involved and observables for FF and QF can overlap significantly. In collaboration with the Australian National University (ANU), Canberra, Australia, several experiments have been carried out in recent years at ANU's Heavy Ion Accelerator Facility (HIAF). Here, a large range of beam-target configurations was examined in order to investigate how QF/FF competition evolves with changes in entrance channels, taking advantage of the wide angular range available for fission-fragment detection with the CUBE detector setup. These data, together with data on fusion-evaporation reactions (e.g., measured at TASCA and SHIP), provide a comprehensive dataset for describing the QF and FF [Khu17]. In particular, measurements of varying QF probabilities in the $^{48}$Ti+$^{204,208}$Pb and $^{50}$Ti+$^{206,208}$Pb reactions have shed light on the strong influence of nuclear structure on the fusion process. Observed differences for $^{48}$Ti+$^{208}$Pb and $^{50}$Ti+$^{206}$Pb, providing distinct experimental evidence that even reactions that form the same compound nucleus and share the same charge product ($Z_p \cdot Z_t$) can exhibit different levels of QF, cannot be explained by current theories. In addition, data collected using beams of $^{48}$Ca, $^{50}$Ti, $^{54}$Cr, $^{58}$Fe and $^{64}$Ni incident on actinide targets has yielded a clear systematic picture of a strong dynamical evolution when moving from $^{48}$Ca to heavier beams. The data are currently under final analysis. To obtain a conclusive picture concerning the choice of the most preferable beam-target combination for the synthesis of new elements beyond $Z$=118, fusion-evaporation cross-section measurements of $^{50}$Ti-induced reactions on any actinide is suggested. Such measurements are planned to be performed, e.g., at RIKEN, Japan and Dubna, Russia.

## *Nuclear Structure*

Construction of the novel ALpha-BEta-GAmma (ALBEGA) multi-coincidence spectroscopy setup for chemically separated samples [DiN15] continued. Advanced prototypes of the two ALBEGA core detectors, whose inner surfaces are covered with a thin Al and $SiO_2$ layer, respectively, have been tested. They were developed to provide i) improved energy resolution and sensitivity to low energy signals produced by electrons, ii) higher photon detection efficiency, and iii) better mechanical stability against pressure differences. The tests performed with several radioactive sources including $^{241}$Am (α particles) and $^{133}$Ba (conversion electrons) confirmed performance according to specifications, including the uniformity and desired very thin thickness of the dead layers, as well as the sensitivity to the low energy signals. In a next step, the detectors will be mounted in a sandwich configuration to produce the final version of the ALBEGA core detector that will be characterized in 2018.

Nuclear decay spectroscopy data collected in parasitic $^{48}$Ca beamtime was examined as part of the commissioning of the new Compact Decay Spectroscopy Setup (COMPASS) [Ack18] detection system at SHIP. Initially, the upgrade of using a higher granularity implantation

detector in the form of a double-sided silicon strip detector was assessed, with long correlation times from the decay $^{254}$No→$^{250}$Fm (~30 min) →$^{246}$Cf measured [Mis18]. In addition, the ability to perform α-γ coincidences was successful, with excited states in $^{249}$Fm populated following the decay of $^{253}$No. Secondly, with the use of new digital electronics (FEBEX3A) [Hof12], the heavy neutron deficient region around *Z*=92-94 was explored. The use of such a fast timing system (20 ns time resolution) enables full chains to be acquired from a region of fast decaying nuclei, where with a conventional analogue system chain members could not be recorded due to dead time of the electronics system. A successful product of this investigation was synthesis of the previously unconfirmed isotope $^{225}$Np. Development work on improvements to the detector system included upgrading to FEBEX4A (10 ns timing resolution) and enhancements to the escape 'box' detectors to maximize efficiency.

## *Mass measurements for nuclear structure studies*

The nuclear structure studies by direct mass spectrometry with SHIPTRAP will be further extended to heavier and more exotic nuclides in the next beamtime period at GSI in 2018. Facing the challenge of ever-lower production rates, it has been worked on measures for a further increase in efficiency and sensitivity. To this end in the recent years, a cryogenic gas cell for SHIPTRAP has recently been built. In the commissioning beamtime in 2015 an increase of the overall efficiency by almost one order of magnitude was achieved [Kal15]. In 2017, additional offline studies were performed with radioactive sources. We studied the extraction of different elements from the gas cell and to optimize the transport of ions to the SHIPTRAP Penning traps following the 2016 relocation of the complete setup [Gia17]. The phase imaging (PI-ICR) measurement technique [Eli13] was further optimized at SHIPTRAP studying systematic uncertainties, exploring the achievable mass resolving power and improving the long-term stability for measurements with lowest yield. The PI-ICR technique has become the new standard for on-line mass measurements of radionuclides worldwide. However, challenges for direct mass spectrometry of the heaviest elements remain. In the next measurement campaign at GSI the identification of low-lying isomeric states in No-, Lr-, and Rf-isotopes is planned. In this mass region the various types of isomeric states are known, many of which have low excitation energies and similar half-lives to the ground state rendering their identification by means of conventional nuclear spectroscopy difficult. The high mass resolving power of the PI-ICR method makes it an ideal choice for this. A mass resolving power of about 100,000 for only 100 ms measurement time has also been reached for heavy ions.

## *Atomic Physics*

The pioneering laser spectroscopy work on nobelium performed at the GSI in recent years led to the observation of more than 30 atomic states in the nobelium atom, among them several Rydberg states [Laa16]. From the convergence of the observed Rydberg series, we eventually obtained the ionization potential based on a two-step laser ionization scheme with high accuracy. However, the presence of buffer gas collisions led to the population of long-lived metastable states below the $^1P_1$ state that was directly excited by the first-step laser. The second laser excitation had sufficient energy to ionize the nobelium atoms from either of the two states. Thus, the identification of the different series was crucial for the IP determination. This was accomplished based on the delayed ionization signal obtained when the second step laser was delayed compared to the first one. In the case of the $^1P_1$ state that features a lifetime of only about 2 ns this signal decayed rapidly, whereas a longer-lived component with a lifetime of tens of ns was observed indicating the population of the lower-lying metastable state. In 2017, the data analysis was completed, and a rate equation model was developed that allowed us to describe the experimental data for nobelium as well as corresponding data for the homolog ytterbium perfectly [Chh17]. Consequently, the first ionization potential of nobelium was determined two orders of magnitude more accurately than before [Chh18].

The analysis of the hyperfine spectroscopy data for $^{253}$No was supported by three different sets of atomic structure calculations that provided the mass shift and field shift constants as well as the hyperfine parameters. The results support the ground state spin and parity assignment of $9/2^-$, previously derived from in-beam gamma spectroscopy, and furthermore provide the magnetic and quadrupole moment of this odd-*A* nucleus accurately. Recently, state-of-the-art density functional calculations reproduced the differential charge radii of lighter actinides [Mar14, Rei17]. These calculations are also in excellent agreement with the nobelium data on differential charge radii. This confirms the theoretical prediction of maximum deformation in the nobelium isotopic chain around neutron number $N$ = 152. It also substantiates the claim of a sizeable central depression in the proton distribution of $^{254}$No, a feature that is only found in superheavy nuclei and originates from their strong Coulomb repulsion. The hyperfine spectroscopy results have been submitted for publication [Rae18].

## *Chemistry*

In the past beamtime periods, experiments to study the chemical behavior of Fl and Nh in comparison with Hg, Tl, Pb, and Rn were performed at TASCA [Blo16]. In addition, the volatility and reactivity of these elements towards surfaces like $SiO_2$ and Au were measured. The comprehensive analysis of these results was continued. For Nh, experimental results [Blo16, Aks17] indicate a reduced volatility and enhanced reactivity compared to Cn and Fl. To render assistance to these gas-phase experiments, calculations of the adsorption energies of these elements and their lighter homologs on a Au(111) surface have been performed using a periodic ADF BAND code. Such periodic calculations of adsorption energies have been performed for the first time for superheavy element systems adsorbing on gold. The results have shown that Cn should indeed be the most volatile element out of

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those under consideration. In addition, Fl should interact with gold at room temperature. Nh, should very strongly interact with gold. Such a different adsorption behavior allows for a good separation between all of these elements using a combination of quartz and gold surfaces [Per17a]. In addition, molecular properties of group-13 hydroxyls (of Tl and Nh) needed for predictions of their reactivity with quartz and gold have been calculated with the use of most advanced relativistic methods. In difference to the conclusion from the earlier predictions, NhOH is expected to be less volatile than TlOH [Per18]. To study Nh under improved conditions, advanced setups for optimized transport of the element under study to the detection setup are under development also allowing investigation of less volatile species. One such approach involves the direct connection of the existing COMPACT detection array [Yak14] to the TASCA Recoil Transfer Chamber (RTC). Another possibility involves the coupling of COMPACT to a recoil separator by employing a buffer-gas stopping cell instead of a classical RTC. First studies of the extraction efficiency of $^{219}$Rn ions obtained from a $^{223}$Ra recoil ion source installed in the SHIPTRAP buffer gas stopping cell [Neu06] into a COMPACT detector array were measured in off-line experiments at GSI. A first on-line experiment with such a setup was performed at the Cyclotron Institute, Texas A&M University, College Station, USA. The buffer gas stopping cell was coupled to the Momentum Achromat Recoil Spectrometer (MARS) [Fol12]. The isotopes $^{182,183}$Hg and $^{199,200}$At were produced in the reactions $^{40}$Ar + $^{147}$Sm and $^{40}$Ar + $^{165}$Ho, isolated in MARS, thermalized in the buffer gas stopping cell, and extracted into the COMPACT by using electric fields. The final analysis of the obtained extraction efficiencies and transport times is currently ongoing.

The studies related to volatile transition metal carbonyl complexes with short-lived isotopes have been continued. Currently, the method of choice is the isolation of the studied isotopes in a recoil separator, followed by their thermalization in an RTC that is flushed with CO-containing carrier gas [Eve14]. As the fusion-evaporation products of asymmetric fusion reactions needed for the productions of sufficiently long-lived isotopes of the elements of interest like Sg, Bh, and Hs, have a relatively large angular and energy spread, transmission efficiencies through a recoil separator like TASCA or GARIS at RIKEN are rather moderate, around 10-15%. Thus, the overall efficiency for the synthesis of carbonyl complexes in combination with physical preseparation is rather low. Therefore, possibilities for the chemical investigation of these compounds without a physical preseparator are currently explored. First experiments performed at the Tandem accelerator at JAEA Tokai, Japan, suggested that the successful synthesis of Os and W carbonyl complexes is feasible if the thermalization of the evaporation residues is spatially decoupled from the chemical synthesis. In that way it is needed, that the formation of the carbonyl complexes takes place in the absence of the beam, which is a strict requirement [Wan14]. In experiments with fission products at the research reactor TRIGA Mainz, the partial efficiencies for the flush-out transfer of non-volatile products from the thermalization chamber into the chemical synthesis chamber were measured and confirm that such an approach allows obtaining higher efficiencies than with preseparation. To support gas-phase experiments on studies of the stability and volatility of carbonyls of the heaviest elements, calculations of the electronic structures and properties of group-6 carbonyls [Ili17] and group-7 carbonyls, including those of Bh, were performed using the most advanced relativistic quantum-chemical methods (ADF BAND, X2c-DFT, DIRAC). The work considers all possible formation reaction scenarios of the single species that do not exist in macrochemistry. The formation mechanisms have been found and the volatile species that should form at the experimental conditions are suggested. Accordingly, the detailed properties of the $M(CO)_5H$ species (M= Tc, Re and Bh) have been calculated, including radicals $M(CO)_5$. Volatilities of these species and first bond dissociation energies have been predicted [Per17b].

Further activities, including those centered at the Helmholtz Institute Mainz, are described in more detail in the contribution to the Annual Report 2018 of the Helmholtz Institute Mainz [HIM18].

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