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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