4.4 Department SHE Chemistry and SHE Physics

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The activities of the SHE Physics and SHE Chemistry departments in 2018 were first focused on the implementation of facility upgrades during the extended period without UNILAC operation, and then on the 2018 UNILAC beamtime block. Selected experiments of the Superheavy Element Chemistry department were performed abroad at JAEA Tokai, Japan. In the following, the most important research topics are briefly summarized.

Highlights in 2018

High-precision mass measurements

The 2018 beamtime campaign at SHIPTRAP allowed extending direct high-precision mass measurements to more exotic nuclides. Thanks to a cryogenic gas stopping cell that was installed in the extended shutdown in 2017/18, measurements were performed more efficiently giving access to the superheavy nuclide ^{257}Rf (Z=104), representing the first such measurement beyond the end of the actinide series. In addition, using the novel PI-ICR technique, originally developed at SHIPTRAP, the masses of the isotopes ^{251,254m}No and $^{254-256}$Lr were measured with statistical uncertainties on a 10^{-9} level. This was the first time that this technique has been applied under extreme conditions of lowest production rates. Exploiting the high mass resolving power up to 10,000,000, low-lying isomers with tens of keV excitation energies ^{251m}No and ^{254m,255m}Lr were identified unambiguously. Such measurements are presently only possible at SHIPTRAP. The data analysis is close to completion. A particular focus was on the investigation of systematic uncertainties in the PI-ICR method and a first publication is in preparation.

Laser spectroscopy

The analysis of the 2016 laser spectroscopy campaign of nobelium at GSI was completed and the results were published in two papers in Phys. Rev. Lett. [1,2]. From more than 30 identified atomic states in nobelium, its first ionization potential was determined with high accuracy. Hyperfine spectroscopy of ^{253}No confirmed a ground state spin parity of $9/2^+$ and provided nuclear model-independent values for the quadrupole moment and the magnetic moment. In addition, differential charge radii for $^{252-254}$No were obtained. The results were in excellent agreement with theoretical work, which accompanied the experimental efforts. The laser spectroscopy on nobelium isotopes and the search for atomic levels in the heavier element lawrencium is scheduled for the 2019/2020 beamtime campaign.

A new setup for high-resolution laser spectroscopy using an in-gas jet approach originally developed by KU Leuven has recently been built in Mainz. The online commissioning at GSI is foreseen to start in 2019 with experiments on the radioisotope ^{155}Yb. An experiment to determine the structure of the 8^- K-isomer in ^{254m}No has been approved by the GSI PAC and is planned to be performed in 2020.

Nuclear reactions

A focus was on the study of nuclear reactions which do not lead to fusion. In a first paper, results from a detailed analysis of reaction products detected during the ^{50}Ti+^{249}Cf irradiation performed in 2012 were published, which yielded information on properties of non-fusion reactions in this system [3]. A second paper reported results obtained in collaboration with the Australian National University, Canberra, addressing differing fusion probabilities in the ^{48}Ti + ^{204,208}Pb and the ^{50}Ti + ^{206,208}Pb reactions [4].

Nuclear structure

TASCA underwent several upgrade measures in 2018 and was thereafter commissioned with parasitic beam; this was used to simultaneously perform some physics measurements, e.g., on ^{250}No or the hitherto unknown ^{244}Md. For this, the updated TASCA focal plane detection systems with full digital electronics on Febex4-basis, which is a development of the Experiment Electronics Laboratory at GSI, was used. The data analysis is close to completion and a first publication is in preparation.

Besides preparations for the approved nuclear spectroscopy experiment on Fl and its daughters, analysis of the data set acquired in 2012 with an emphasis on "unwanted" background products was continued, and refined data on ^{219}Ra and ^{215}Rn was obtained by the collaborators from Lund University, Sweden [5]. At SHIP the commissioning of the new focal plane detector setup COMPASS was completed [6]. This system comprises a compact box geometry with pixelized silicon strip detectors that can be surrounded by Ge detectors for efficient alpha-gamma spectroscopy. Further improvements of the SHIP decay spectroscopy setup have been tested in the 2018 UNILAC engineering run. First experiments with COMPASS on neutron-deficient Np and Pa isotopes have been performed by Lund University, and the results will soon be published.

SHE Chemical Properties

In past beamtimes, experiments on the chemical behaviour of Fl and Nh in comparison with Hg, Tl, Pb, and Rn were performed at TASCA, and a paper focusing on the lighter homologs was published in 2018 [7]. The focus of the next experiments is on Nh, member of group 13 and a homolog of Tl. To lend assistance to the current suite of gas-phase chromatography experiments at TASCA on the volatility and reactivity of Nh. With the Amsterdam Density Functionale ADF calculations of adsorption energies of group-13 hydroxyls (of Tl and Nh) on gold were performed and published [8,9]. The results have shown that the trend in adsorption of MOH, TlOH < NhOH, is opposite to the one of the atoms, where Tl > Nh. The adsorption enthalpies of Nh and NhOH were found to be very similar, so that the identification of the chemical composition by measuring adsorption temperature will be difficult. Experimental work focused on establishing the optimum experimental setup and operation parameters for the main experiment on Nh chemistry, which is scheduled to take place at the UNILAC at GSI in 2020. For this, the combination of an improved version of the gas phase chromatography setup COMPACT, termed miniCOMPACT and adapted to short-lived more reactive species, with the TASCA separator has been optimized, studying different coupling options including a traditional gas-filled Recoil Transfer Chamber (RTC) as well as the previous SHIPTRAP buffer-gas stopping cell equipped with DC gradient and RF repeller funnel. In online experiments the short-lived isotopes ^{204,205}Fr, produced via the reaction ^{40}Ar + ^{169}Tm, were successfully extracted from the RTC into miniCOMPACT. The experiments with the RTC-miniCOMPACT combination were carried out in the 2018 beamtime period, while those with the buffer-gas-cell/miniCOMPACT combination are foreseen for the 2019 beamtime period. In preparation of the latter, off-line measurements were performed with ^{223}Ra and ^{225}Ac recoil ion sources installed in the buffer gas stopping cell. In separate experiments, the gas-catcher-COMPACT-setup as well as the new RTC-miniCOMPACT setup were flushed with helium gas and allowed performing gas phase chromatography studies the recoil ions ^{219}Rn ($T_{1/2}$ = 3.96 s) or ^{217}At ($T_{1/2}$ = 32.3 ms). The efficiency for transporting these isotopes from the source to the COMPACT detector as well as the transport time were studied.

The studies of volatile transition metal carbonyl complexes with short-lived isotopes has been continued, using the two-chamber technique which was developed last year. Building up on the successful experiments with fission products at TRIGA Mainz, which focused on studying and optimizing the partial efficiency of the flushing-out the first into the second chamber, an on-line experiment with the heavier homologs was carried out at JAEA Tokai. This confirmed that such an approach allows obtaining higher efficiencies than with preseparation, however, rather long-lived isotopes were used to minimize the influence of kinetic effects. The latter will be studied in coming experiments with short-lived alpha-decaying isotopes, for which beamtime is approved at JAEA Tokai as well as at RIKEN, Wako. With the aim to support gas-phase experiments on study of stability and volatility of carbonyls of the heaviest elements, calculations of the electronic structures and properties of group-7 carbonyls, including those of Bh, were published [10].The most advanced relativistic quantum-chemical methods (ADF BAND, X2c-DFT, DIRAC) were utilized, and properties of the $M(CO)_5H$ species (M= Tc, Re and Bh) have been determined, including radicals $M(CO)_5$. Volatility of these species and first bond dissociation energies were predicted.

Outlook for 2019

The UNILAC beamtime in spring 2019 will be the main activity with a focus on laser spectroscopy of nobelium and lawrencium at SHIP, on the spectroscopy of flerovium decay chains at TASCA, and parasitic beamtimes at both devices to perform parameter studies, e.g., in preparation of the approved beamtime on nihonium chemical properties planned for 2020.

Selected publications of 2018

- [1] Raeder, S. ; Ackermann, D. ; Backe, H. ; et al.: P**robing Sizes and Shapes of Nobelium Isotopes by Laser Spectroscopy**. *Physical review letters 120(23),* 232503 (2018) DOI:10.1103/PhysRevLett.120.232503
- [2] Chhetri, P. ; Ackermann, D. ; Backe, H. ; et al.: **Precision Measurement of the First Ionization Potential of Nobelium.** *Physical review letters 120(26),* 263003 (2018) DOI:10.1103/PhysRevLett.120.263003
- [3] Di Nitto, A. ; Khuyagbaatar, J. ; Ackermann, D. ; et al.: **Study of non-fusion products in the ^{50}Ti + ^{249}Cf reaction.** *Physics letters / B B 784*, 199 - 205 (2018) DOI:10.1016/j.physletb.2018.07.058
- [4] Khuyagbaatar, J. ; David, H. M. ; Hinde, D. J. ; et al.: **Nuclear structure dependence of fusion hindrance in heavy element synthesis.** *Physical review / C 97(6)*, 064618 (2018) DOI:10.1103/PhysRevC.97.064618
- [5] Såmark-Roth, A. ; Sarmiento, L. G. ; Rudolph, D. ; et al.: **Low-lying states in ^{219}Ra and ^{215}Rn : Sampling microsecond -decaying nuclei.** *Physical review / C covering nuclear physics 98(4),* 044307 (2018) DOI:10.1103/PhysRevC.98.044307
- [6] Ackermann, D. ; Mistry, A. K. ; Heßberger, F. ; et al.: **COMPASS—A COMPAct decay spectroscopy set-up.** *Nuclear instruments & methods in physics research / A Accelerators, spectrometers, detectors and associated equipment Section A 907*, 81 - 89 (2018) DOI:10.1016/j.nima.2018.01.096
- [7] Lens, L. ; Yakushev, A. ; Düllmann, C. E. ; et al.**: Online chemical adsorption studies of Hg, Tl, and Pb on SiO_2 and Au surfaces in preparation for chemical investigations on Cn, Nh, and Fl at TASCAKC**. *Radiochimica acta 2017-2914(0),* 1-14 (2018) DOI:10.1515/ract-2017-2914
- [8] Pershina, V. ; Iliaš, M.: **Electronic structure and properties of MAu and MOH, where M = Tl and Nh: New data.** *Chemical physics letters 694,* 107 - 111 (2018) DOI: 10.1016/j.cplett.2018.01.045
- [9] Pershina, V.: **Reactivity of Superheavy Elements Cn, Nh, and Fl and Their Lighter Homologues Hg, Tl, and Pb, Respectively, with a Gold Surface from Periodic DFT Calculations.** *Inorganic chemistry 57(7),* 3948 - 3955 (2018) DOI:10.1021/acs.inorgchem.8b00101
- [10] Pershina, V. ; Iliaš, M.: **Carbonyl compounds of Tc, Re, and Bh: Electronic structure, bonding, and volatility.** T*he journal of chemical physics 149(20),* 204306 (2018) DOI:10.1063/1.5055066