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4.4 SHE departments and HIM SHE section

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The SHE departments devoted to the research of superheavy elements, operate the recoil separators SHIP and TASCA and their ancillary installations including SHIPTRAP and a laser spectroscopy setup at SHIP as well as chemistry and nuclear spectroscopy setups at TASCA. In 2019, the activities at GSI focused on the UNILAC beamtime within the FAIR Phase-0 program and on the analysis of data obtained in prior beamtimes. At HIM, the advancement of actinide sample preparation, manipulation, and characterization for various applications was most central. In addition, technical developments, for example for single-ion mass measurements, have been performed.

Synthesis/nuclear reactions

The six SHE with atomic numbers Z=107-112 have been discovered in cold fusion reactions of heavy projectiles in the range of Cr-Zn with lead/bismuth targets. Cross sections to form SHE in cold fusion reactions decrease drastically with increasing Z of the compound nucleus. The dynamical mechanisms underlying this trend remained only partially understood. Experiments aiming at further elucidating cold fusion reaction cross section trends were carried out in collaborative work of the SHE Chemistry departments at GSI and HIM and the nuclear reactions group at the Australian National University (ANU), Canberra, Australia, at the Heavy Ion Accelerator Facility of ANU. The results [1] show that cold fusion reactions occur under a strong dynamical regime, disagreeing with a commonly held assumption that thermal-diffusion-like processes play a dominant role. This calls for microscopic approaches to describe this reaction step, hence stimulating further theoretical work.

Recently, complete fusion reactions involving proton emission during deexcitation of the compound nucleus have been suggested for production of SHE isotopes not accessible by neutron evaporation channels. At FLNR/JINR Dubna a cross section of 4 pb for the p-channel was measured at an excitation energy of E* = 29 MeV in bombardments of ^{209}Bi with ^{50}Ti [2]. At SHIP the maximum of that channel was observed at lower energy E* = 21 MeV in a first experiment in 1985 [3]. This observation was confirmed by a reanalysis of all irradiations of ^{209}Bi with ^{50}Ti performed at SHIP from 1997 to 2014 that resulted in a cross section maximum of 18 +-8 pb at E* = 21 MeV for the exotic p – deexcitation channel [4].

Nuclear structure studies

Motivated by the quest for a magic proton number Z = 114 (flerovium Fl) and the search for even-Z odd-N experimental anchor points relevant for nuclear structure theory in the superheavy element regime, a first attempt was conducted to explore the nuclear reactions ^{48}Ca+^{242}Pu and ^{48}Ca+^{244}Pu using high-resolution alpha-photon coincidence spectroscopy along ^{287,289}Fl decay chains behind the gas-filled separator TASCA. The upgraded TASISpec decay station, contributed from Lund University, Sweden, was used, employing new dedicated Compex germanium detector modules [5] financed by the Knut and Alice Wallenberg foundation. First, the reaction ^{48}Ca+^{206,207}Pb served to verify experimental parameters like the TASCA transmission and target-segment to α-decay correlations. Short-lived nuclei were produced via ^{48}Ca+^{nat}Hf to investigate pile-up events [6].

The set-up was found working flawlessly during the main experiment. The first 12 of the approved 25 days were scheduled in 2019; a reduced available beam-intensity led to the collected beam integral being equivalent to only about 6-7 effective days. Nevertheless, nine Fl decay chains were identified, corresponding to a production cross section of approximately 10 pb, a value which is in agreement with literature.

In further TASCA experiments, the sampling ADC "FEBEX" modules developed by the GSI's Experiment Electronics department have been extensively employed, e.g., to detect conversion electrons from the deexcitation of isomeric states, e.g., in ^{254,255}Rf [7]. This demonstrates the potential of fast digital electronics for measurements of isomeric states in the heaviest nuclei, which can only be produced in small quantities. The technique also aided in studies of the two known short-lived fission activities associated with ^{250}No - one of them has been assigned to a K-isomer - performed at the gas-filled recoil separator RITU at the University of Jyväskylä, Finland [8]. By observing conversion electrons preceding fission of one of those activities, the ground- and isomeric-state assignments could be made.

The probability of the electron capture delayed fission process (ECDF), which is known to occur in 29 nuclei with Z = 81-101, has so far not been described satisfactorily, especially in superheavy nuclei. A semi-empirical estimate of this process was developed, which accounts for all known cases [6]. The delayed fission appears to be strongly affected by the shape of the excited nucleus, in which the fission path towards scission is different than that of the nucleus in its ground state. In [6], semi-empirical predictions on the ECDF probabilities in odd-odd nuclei with 79 < Z < 119 are given. According to these estimates, beta-delayed fission decay modes appear to be the among the main decay modes in superheavy nuclei which undergo beta decay.

The results of a detailed alpha-conversion electron-gamma spectroscopy study of ^{258}Db, produced at SHIP in the ^{209}Bi(^{50}Ti, 1n)^{258}Db reaction, and its α-decay daughter products were published [10]. Two long-lived α-decaying states in ^{258}Db were observed and tentative spin-parity assignments were made for the ground state and the isomeric state, which was measured at an excitation energy of 51 keV. The α-decay daughter isotope ^{254}Lr displays a similar behavior. The isomeric level of this isotope was placed at 108 keV. Two long-lived α-decaying states were observed in the granddaughter ^{250}Md, also featuring an isomeric state positioned at an excitation energy of 123 keV. Partial, tentative level schemes for ^{254}Lr, ^{250}Md and ^{246}Es were proposed based on the α-γ coincidences.

Neutron deficient U, Pa, and Np isotopes with neutron numbers around N = 126 were investigated at SHIP using the COMPASS detector setup [8]. Studying the decay of these often short-lived isotopes benefits from the use of fast digital electronics. Besides improved data on decay properties, the cross sections for the production of these isotopes with ^{48}Ca were determined, showing that ^{48}Ca does not provide an advantage as a projectile to access this region of the nuclear chart.

Nuclear properties from laser spectroscopy

The laser spectroscopy experiments at SHIP were extended to ^{255}No. The isotope shift and the hyperfine splitting of the $^1S_1$-$^1P_1$ transition were measured employing the RADRIS method established in pioneering experiments on $^{252\text{-}254}$No. ^{255}No was produced via the electron capture decay of ^{255}Lr with a yield of about 0.05 atoms per second. This production method was necessary due to the close-lying alpha energies of ^{254}No and ^{255}No that prevented an unambiguous identification in a direct production. Despite the low yield a fully resolved hyperfine spectrum of ^{255}No was obtained confirming a nuclear spin of ½. The data is still under analysis and will provide the magnetic moment of ^{255}No. The mean-square charge radius derived from the isotope shift extends the systematics in the nobelium isotope chain beyond the N=152 shell closure. The achieved sensitivity opens the door for future experiments on Lr (Z=103) for which preparatory studies identifying the optimum filament material for efficient desorption were carried out [12].

The accurate determination of nuclear properties by hyperfine spectroscopy in heavy elements calls for a high spectral resolution. In the RADRIS technique, the buffer gas environment results in a Doppler broadening of few GHz. This prevents resolving individual hyperfine components in some cases. A new approach, in which atoms are extracted in a supersonic gas jet and then laser

ionized perpendicular to this jet, reduces the Doppler broadening by about one order of magnitude without sacrificing efficiency. At HIM, a novel setup was constructed in collaboration with KU Leuven to implement this method at GSI utilizing the filament technique successfully applied in nobelium laser spectroscopy [13]. The new method will extend the reach towards short-lived isotopes not accessible with the RADRIS technique. For example, it will facilitate a measurement of the magnetic moment of the K-isomer in ^{254}No to unambiguously assign its nuclear configuration. The commissioning of the gas jet setup is ongoing and a first online test with ^{155}Yb was completed in 2019.

The activities on the laser spectroscopy were complemented by an off-line measurement campaign on long-lived Es and Fm isotopes which became available in nano- and picogramm quantities. Here, a first set of measurements allowed for a detailed investigation of the atomic structure of einsteinium.

Nuclear properties from mass measurements

Several technical developments for extending the reach of high-precision mass measurements have been advanced. A new low-noise detection circuit based on a quartz resonator for the Fourier-Transform Ion-Cyclotron-Resonance (FT-ICR) detection technique has been implemented at TRIGA-TRAP [14]. The device is rather compact and relies on commercial quartzes that feature a high quality factor at room temperature. A proof-of-principle mass measurement on ^{206,207}Pb ions was performed demonstrating a sensitivity on the order of tens of ions for room-temperature operation. The results are relevant for the design of tailored FT-ICR systems for SHIPTRAP and for the future MATS setup at FAIR.

Another novel detection technique for single-ion mass measurements with optical detection is being developed at the University of Granada within the TRAPSENSOR project [15]. An exotic ion of interest will be trapped in a laser-cooled ion crystal in a Penning trap. The sensor ion ^{40}Ca was efficiently Doppler laser cooled in the strong magnetic field of a Penning trap using 12 laser beams. Once a two-ion crystal cooled to the (quantum mechanical) ground state has been realized, mass measurements can be performed applying quantum logic techniques.

For future off-line measurements of long-lived nuclides, the laser ablation ion source of SHIPTRAP has been improved by an optimized miniature radiofrequency quadrupole design. This will facilitate measurements with lower atom numbers on the order of 10^{14} and below. The characterization and optimization of the cryogenic gas cell for SHIPTRAP was completed [16]. The improved performance enables future mass measurements of Rf and Db isotopes. The data analysis of the 2018 SHIPTRAP campaign was completed and systematic uncertainties to be considered in low-rate measurements with the phase-imaging technique were investigated. A publication is in preparation.

Chemical properties

The evaluation of all available experimental data on the chemical interaction of Fl (element 114) with a gold surface has been finalized. A detailed analysis of a thin gold layer on a detector surface showed this surface to be inhomogeneous. An extended model for the interaction of Fl with such surfaces including the mobility of an adsorbed atom on the surface, was proposed. This model allows describing all available experimental data well and reveals that the strength of the interaction is decisively site-dependent. A publication detailing the chemical properties of Fl has been submitted.

Experimentally, the focus of experiments at GSI was on preparing and optimizing the experimental setup for the study of chemical properties of nihonium Nh (element 113). The first chemical study of Nh at TASCA was attempted in 2016. No Nh events were observed in a three-week experiment. Since then, several preparatory studies with the lighter homolog Tl as well as further model elements have been performed, aiming at a faster and more efficient extraction of short-lived and chemically reactive species into the gas chromatography and detection setup COMPACT. In the 2019 beamtime, reactive $^{205\text{-}207}$Fr was successfully extracted into a new miniaturized version

(miniCOMPACT), which was used in a combination with i) a modified recoil transfer chamber, and ii) with the former SHIP-TRAP gas buffer cell (BGC). Based on the results obtained especially with approach i), a four-week long Nh chemistry experiment is scheduled at GSI for 2020. The 2019 GSI experiments employing approach ii) proved that the combination of such a cell with the COMPACT setup is technically feasible. After first proof-of-principle experiments performed off-line using ^{219}Rn ions and ^{221}Fr ions from radioactive recoil ion sources, and a first beam-test performed at Texas A&M University in 2017, the main focus was on quantitatively confirming the extraction efficiencies achieved in off-line studies. The extraction efficiencies obtained off-line were confirmed by these experiments using the ^{165}Ho(^{48}Ca,xn)$^{213-n}$Fr, ^{165}Ho(^{40}Ar,xn)$^{205-n}$At, and ^{144,147}Sm(^{40}Ar,xn)$^{187-n}$Hg reactions, opening the prospects for chemistry experiments with very short-lived SHE isotopes beyond Fl. To overcome limitations of the current BGC, a novel design was worked out [17].

Studies of molecular chemical systems with SHE focused on further developments of the carbonyl chemical system, with the aim of a yield increase to open up perspectives to go beyond $Sg(CO)_6$. A novel technique omitting the recoil separator step to spatially decouple recoil ion thermalization and gas phase chemical synthesis and replace it by a system comprising two separate, but directly coupled chambers for each of these steps has been developed. This was successfully tested with short-lived transition metal isotopes produced via ^{235}U(n,f) at the research reactor TRIGA Mainz and via ^{248}Cm(sf); fission products, stopped inside a gas volume directly behind the target, were transported to a second, directly connected chamber. There, the products were either collected for direct counting to allow for yield measurements, or converted into volatile carbonyl complexes and transported to remote counting setups. Results from a beamtime conducted at the Tandem accelerator at JAEA Tokai, in which this approach was applied to 5d elements produced in ^{19}F induced reactions were evaluated, and confirmed that the two-chamber approach indeed allows synthesizing carbonyl complexes of accelerator-produced single transition metal isotopes with higher overall efficiency than the previously used preseparator approach.

With the aim to support such gas-phase experiments on the volatility of Nh with respect to its homolog Tl, predictions of adsorption behaviour of group-13 hydroxides on a gold surface were made using periodic density functional theory (DFT) codes [18]. It was shown that NhOH should adsorb relatively well on gold, almost as strongly as elemental Nh. This similarity will render the identification of the chemical species difficult. Further work focused on the carbonyl chemical system. The program of the SHE section was supported by an extended visit of Professor M. Iliaš from Matej Bel University, Banská Bystrica, Slovakia via the HIM Fellow Program. With the aim to establish a chemical character of Lr upon adsorption of various surfaces of Ta, periodic ADF BAND calculations were performed for the Ta bulk and adsorption energies of Lu, Lr and Tl. Adsorption of Lr was shown to be similar to that of Lu, but different than that of Tl, showing Lr to be a good homolog of Lu. Its 6d and 7s atomic orbitals are involved in bonding. This was done in collaboration with JAEA Tokai, Japan, where preliminary experimental results for Lr were obtained; these agree very well with the calculations. With the aim to predict formation reactions of Mc and Mc^+ in chemistry experiments, various formation reactions in the gas-phase were considered using the most accurate relativistic methods.

Laser-produced protons inducing nuclear reactions at PHELIX

The first experiment on study of fission induced by laser-beam generated protons was conducted at PHELIX. Protons accelerated by a laser pulse, penetrated a Ti window and impinged on a 25-µm thick U foil, inducing fission. Fission products recoiling from the foil were stopped in a gas-filled chamber. Using an inert-gas jet transport technique, volatile fission products (Xe and I isotopes) re-coiling were transported to a collector and identified via gamma spectroscopy. The SHE chemistry department contributed on the gas jet transport and identification aspects of this experiment.

Tailor-made (actinide) samples

A main activity in Mainz is the production of tailor-made samples of (radio)isotopes for applications in basic chemical and physics research. These activities expanded into several areas.

Fundamental work to improve target production for SHE via the well-established "molecular plating" (MP) method was performed and novel electrochemical strategies were explored, not least to establish the capability to produce metallic layers that have a more favorable ratio of desired (=actinide) to undesired (=all other elements) atoms, even after oxidation, which occurs rapidly at air. The lanthanides and actinides are very electropositive elements. The metallic f-elements react with water to form hydrogen and metal hydroxides or oxides. Therefore, the pure metals cannot be deposited electrochemically from an aqueous solution. Nonetheless, in MP, the lanthanides and actinides are added in aqueous nitric acid. The incorporation of aqueous nitrates has several disadvantages, such as the gas development during the MP process. In f-metal chemistry, established alternatives to the nitrate salts include water-free tosylate or triflate salts. Both classes of substances can be easily synthesized and are adequately described in the literature. Initial tests with tosylate have already shown a significant improvement in thin-film quality. ^{248}Cm targets produced by this method were successfully used in the muX-experiment at Paul-Scherrer Institute in Villigen, Switzerland, where conventional MP-targets have failed before. As a next step, irradiation test of triflate targets will be carried out at GSI to explore the suitability of such targets for heavy-ion research.

^{233}U recoil ion sources providing the alpha-decay daughter ^{229m}Th, which were prepared at HIM, were employed by collaboration partners at LMU Munich in experiments to determine the excitation energy of this exotic nuclear isomeric state. It is the by far lowest-lying known such state in any nucleus, located at an energy below 10 eV. Using the ground state ^{229}Th, laser light provides sufficient energy to bridge these 10 eV, rendering the ^{229m}Th system the only known case where laser techniques are applicable to manipulate a nuclear system. This is conjectured to provide the basis for a future "nuclear clock", potentially outperforming the most precise current atomic clocks owing to the enhanced shielding of the oscillating system, i.e., the nucleus, by the electron shell. The excitation energy, though, has so far only been inferred indirectly. Using a retarding field electron spectrometer, the energy of electrons emitted in the deexcitation of the isomer to the ground state was measured, thus giving experimental information on the excitation energy, which is 8.28(17) eV, corresponding to a wavelength of 149.7(31) nm [19]. This paves the way for the development of suitable laser systems to determine the exact value of the excitation energy with a precision typical of laser-based approaches and is thus a milestone on the way to the nuclear clock. Scientists from LMU Munich, MPIK Heidelberg, TU Vienna, U. Bonn, GSI Darmstadt, HIM Mainz and JGU Mainz contributed to this achievement.

A multitude of further targets and sources, mostly of actinides were supplied as a contribution to collaborative work, including various ^{229}Th and ^{233}U sources for studies connected to ^{229m}Th, e.g., for UCLA, CA; USA; JILA Boulder, CA, USA; Univ. Heidelberg, Germany; LMU Munich, Germany; PTB Braunschweig, Germany; Univ. Jyvaskyla, Finland; as well as samples of Am, Cm, and Cf for SOREQ, Yavne, Israel, and for laser spectroscopy studies at Institute of Physics in Mainz.

Trapping And sympathetic Cooling of Thorium Ions with Calcium (TACTICa)

The TACTICa Collaboration is working on thorium ion spectroscopy, especially of the ^{229m}Th in a variety of charge states, for new insights in fundamental physics. The thorium ions will be loaded into a linear Paul trap and sympathetically cooled in a calcium-ion crystal. For a successful loading, the ions need to have a kinetic energy below 1 keV. In case of the $^{229(m)}$Th, the ions are produced by α decay of ^{233}U with a recoil energy of 84 keV. This kinetic energy is reduced by electrostatic deceleration to retain the initial charge state distribution of the Th ions. Furthermore, an ideal recoil ion source needs to be fabricated consisting of a single layer of uranium atoms to guarantee both, a high recoil efficiency and a negligible deceleration of the ions by interactions with the source material. Several fabrication methods were investigated for the potential of producing uranium monolayers. These include MP, Drop-on-Demand ink-jet printing, chelation by functionalized silicon surfaces and self-adsorption on thermally oxidized titanium foils. The α spectra of sources produced by these methods were qualitatively characterized and compared with advanced alpha spectrometric simulations, to gain insights about the deposited source composition. The investigations lead to fabrication of high qualitative recoil ion sources. The design of the TACTICa source section was finished [20], including IBSIMU ion flight simulations of the decelerated thorium recoil ions. The extraction efficiency is expected to be near to the geometric efficiency of about 2×10^{-5} and therefore a rate of about 1 ion/min is expected. The recoil ion source will be connected with a laser ablation ion source via an electrostatic quadrupole bender, therefore giving access to six iso-

topes of thorium for loading into the Paul trap. A Wien filter will be used to investigate the charge distribution of Th recoil ions and to select a specific charge state for loading into the Paul trap.

An Offline Deposit Irradiation (ODIn) device at HIM Mainz

The ODIn pilot setup [21], comprising a sample irradiation position and two low-energy particle sources, i.e., an electron gun providing electron beams with energies of 10 eV up to 1.5 keV at currents in the 1 nA up to 100 µA range, and a sputter ion gun based on microwave plasma discharge providing beams of gaseous elements at energies of up to 5 keV with currents up to 2 mA, was commissioned. This will serve to investigate to which extent it may be possible to perform the online baking-in process, traditionally tied to heavy-ion accelerator facilities, also at much smaller, accelerator-independent setups. For comparison MP produced lead targets were irradiated at ODIn and at TASCA (5 MeV/u ^{48}Ca; I=410^{12} Ion/s). They showed initially a similar chemical conversion from lead carbonate to lead(II) oxide. With increasing fluences, defect structures in the Raman spectrum could also be detected.

International Year of the Periodic Table and TAN conference in Wilhelmshaven

2019 had been declared the International Year of the Periodic Table (IYPT) by the United Nations to celebrate the 150th anniversary. The SHE section / departments were involved in many activities related to IYPT. One highlight was a special symposium organized within the 6th international TAN conference (TAN 19), chaired by Ch.E. Düllmann and M. Block, with 125 participants form 19 countries. The symposium brought together members of the discovery teams of elements 112-118, including Peter Armbruster, Gottfried Münzenberg, and Yuri Oganessian.

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