

TASCA 16

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15th Workshop on
Recoil Separator for Superheavy Element Chemistry



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Program and Abstracts

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16:05	Theoretical Study of Molecular Properties of Group-VI, VII and VIII Heavy and Superheavy Metal Carbonyls	M. Ilias (GSI / HIM)	9
16:25	Studies of the thermal stability of Sg(CO) ₆	R. Eichler (PSI Villigen)	10
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*15 minutes presentation plus 10 minutes discussion time

**15 minutes presentation plus 5 minutes discussion time



The first ionization potential of nobelium

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A stringent test of modern atomic theories addressing relativistic and quantum electrodynamic effects is provided by investigating spectral lines of the heaviest elements. Albeit the strong efforts undertaken and the large optical spectroscopy research programs initiated in the last century, such studies were deemed to reach a barrier of applicability at the element einsteinium and, more than a decade now, at fermium. Only recently, the first foray of optical spectroscopy into the region of the transfermium elements was reported [1]. Several atomic transitions in the element nobelium were observed and characterized, revealing atomic structure information at production rates of a few atoms per second at most. In addition, an upper-limit value for the first ionization potential (IP) of nobelium was deduced. In this year's beamtime at the GSI, the studies in nobelium were extended, enabling the IP to be extracted at highest precision. Furthermore, first steps towards optical spectroscopy of the element lawrencium were initiated. In this talk, a report on the recent achievements will be given.

References

- [1] M. Laatiaoui, *First observation of an atomic level for the element nobelium*, TASCA15 workshop, GSI, Darmstadt, October 23, 2015; https://www-windows.gsi.de/tasca15/program/contributions/TASCA15_cont_Laatiaoui_2.pdf

Measurements of the first ionization potential of the heaviest actinides

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We successfully measured the first ionization potential (IP_1) of lawrencium (Lr, $Z = 103$) using a surface ionization method [1]. The result suggests that Lr has a loosely-bound electron in the outermost orbital. In contrast to Lr, nobelium (No, $Z = 102$) is expected to have the highest IP_1 among the actinide elements owing to its fully-filled $5f$ and the $7s$ orbitals; due to the similarity of heavy actinides to heavy lanthanides, it is expected that the IP_1 value of heavy actinoids would increase monotonically with filling electrons up in the $5f$ orbital like heavy lanthanoids.

Recently, we have ionized and mass-separated ^{257}No , ^{251}Md and ^{249}Fm , and successfully determined their IP_1 values. The obtained experimental results are in good agreement with the relativistic coupled cluster calculations as well as with earlier predictions.

Details of the experiments and results will be given in the presentation.

References

- [1] T. K. Sato, et al., *Nature* **520** (2015) 209-211



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

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The neutron-deficient ^{240}Es nucleus was produced for the first time using the fusion-evaporation reaction $^{209}\text{Bi}(^{34}\text{S},3n)^{240}\text{Es}$ at the Accelerator Laboratory of University of Jyväskylä (JYFL), Finland. The gas-filled recoil separator RITU [1] was used to separate the fusion-evaporation products from the primary and scattered beam. The radioactive decays originating from ^{240}Es and its daughters were measured with the focal plane spectrometer GREAT [2].

Identification of ^{240}Es was done by detecting recoil correlated α decays feeding to the known granddaughter ^{236}Cm . A significantly high ECDF probability was measured for ^{240}Es [3]. The hitherto unknown ^{236}Bk isotope that was populated in the α decay of ^{240}Es was identified by its ECDF branch [3]. The results on the decay properties of ^{240}Es and ^{236}Bk together with the data analysis will be presented.

References

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- [3] J. Konki et al., to be published

Statistical analysis of decay chains of the heaviest elements

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The heaviest elements have all been discovered through the observation of correlated alpha decay chains. Similarities in the characteristics of different chains, i.e. lifetimes, decay energies and chain lengths, have been used to group the observed chains and assign the groups to different radioactive species. In particular for odd nuclei, the decay energies are difficult to use as the decays often can proceed to various excited states in the daughter nuclei. Neither is the length of a chain necessarily a good descriptor, as fission branches and alpha decay branches can coexist. The measured correlation times in a certain decay step, however, must come from an exponential distribution if the decays come from the same nuclear species. Around the turn of the century, K.-H. Schmidt compared the standard deviation of measured lifetimes within a set of chains with the distributions of standard deviations of Monte Carlo generated sets of chains in order to assess whether the grouping of chains is correct or not [1]. This method deals with each decay step in a chain separately. We have generalized the method to simultaneously take into account all lifetimes along the chains [2]. The method will be outlined and applications of the test to chains assigned to start from element 115 and element 117 will be presented.

References

- [1] H.-K. Schmidt et al., Eur. Phys. J. A **8**, 141 (2000)
- [2] U. Forsberg et al., Nucl. Phys. A **953**, 117 (2016).



Half a century gas phase chemistry in transactinide research

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The potential of gas phase separation to study – and at the beginning also to discover – elements beyond the actinides was first proposed by researchers from Dubna 50 years ago (I. Zvara et al. [1]). Approval of discovery of Rf and Db by IUPAC was – at least partly – based on the chemical studies using thermochromatography of halide molecules with those elements. Drawback of the applied technique was the fact that a fast chemical separation was coupled to an off-line analysis of spontaneously fissioning products. This decay mode is not nuclide specific. This often caused controversial discussions about assignments of separated species. The first fast on-line gas chemical separation coupled to an on-line detector system able to measure α - as well as coincident fragments from SF events was developed at GSI Darmstadt and applied to search for superheavy elements in the reactions of $^{238}\text{U}+^{238}\text{U}^2$, $^{238}\text{U}+^{248}\text{Cm}$ [3] (both in transfer reactions) and $^{48}\text{Ca}+^{248}\text{Cm}$ [4] (in a fusion reaction). This system was called OLGA I (On-Line Gas chemistry Apparatus). Separation times were in the few second range. While OLGA I was designed for inert gas application only a next generation (OLGA II) [5]. was able to separate halides of early transactinides using highly corrosive gases such as HBr, HCl or Cl₂. This system was used to perform chemical studies of Db [6]. The separation time was about half a minute. In order to proceed to even heavier elements, separation times had to be reduced to a few seconds. This was achieved with OLGA III. This set-up was then used for the first ever chemical studies of Sg [7] and Bh [8]. All OLGA systems used isothermal gas chromatography in quartz columns. With element 108 (Hs) thermochromatography was again chosen, but now combined with an online detection system (IVO; On-line Volatilisation and On-line detection). This acronym should honor Ivo Zvara's contribution in the early development of thermochromatography for heaviest element research. Elements Hs (in form of its tetroxide) [9] on quartz surfaces as well as of Cn [10] and Fl [11] (in their elemental state) on gold surfaces were then investigated with IVO techniques by two groups from PSI/FLNR and GSI/Mainz. Most recently a slightly modified version of IVO is applied in ongoing studies of element 113 at Dubna and is used for an ongoing study at GSI [12].

References

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- [4] P. Armbruster et al., Phys. Rev. Lett., **54**, 406 (1985)
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- [12] S.N. Dmitriev et al., Mendeleev Comm., **24**, 233 (2014) & Ch.E. Düllmann, pers. comm. (2016)



THEORETICAL CHEMISTRY OF ELEMENTS 112-114

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A number of important and interesting gas-phase chromatography experiments have been conducted on volatility of superheavy elements (SHE) 112 (Cn) and 114 (Fl) [1-3]. The first attempt of chemical characterization of element 113 has also been announced recently [4]. Accordingly, volatility of these elements and their homologs was studied as adsorption process on a gold surface of detectors of a chromatography column. These SHE were shown to exhibit extremely high volatility that was explained by influence of strong relativistic effects on their electron shells [5-7]. The experiments on Cn and Fl have, however, revealed some disagreement with respect to the chemical conclusions on the nature of the atom-surface interactions and the adsorption enthalpy values [2,3].

Future more elaborated experiments on adsorption behaviour of these elements are supposed to use a silicon oxide surface of the detectors of a chromatography column in addition to gold [8,9]. This should enable one to better distinguish between properties of the homologs and between Cn and Fl, particularly. To render assistance to those demanding experiments with single atoms, our traditional theoretical research has recently been focusing on predictions of interaction of all these elements with a silicon oxide surface, as well as on a more detailed study of their interaction with gold. In difference to the earlier predictions of adsorption of these elements on gold using a cluster approach [7,10,11], the present calculations were performed for the first time using a periodic relativistic code, the ADF BAND [12]. It allows for modelling adsorption processes on extended systems, i.e., on real surfaces of solids. In the talk, results of those investigations aiming at predictions of the adsorption behaviour of Cn, Fl and element 113 along with their homologs on quartz and gold at the experimental temperatures are presented. A dramatic influence of relativistic effects on the properties and experimental behaviour of the heaviest elements is demonstrated

References

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Chemical study of element 113 (nihonium) at GSI

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In the last decade, investigations on the chemistry of superheavy elements (SHE) have been focused on element 112 (Cn) and 114 (Fl). These heavy metals are proven to behave very unusual due to the strongest relativistic effect on their closed electron shells. Recent advanced experiments on Fl at GSI resulted in the unambiguous identification of several decay chains under extremely low background conditions and demonstrated a high efficiency in chemistry experiments with short-lived nuclei ($T_{1/2} \sim 1$ s). Presently, element 113 is on the agenda of chemical studies with SHE. Its properties should be influenced by strong relativistic effects and by one unpaired electron in the valence shell. First chemical studies on element 113 without pre-separation were performed by the FLNR collaboration in Dubna (Russia), where the observation of the adsorption of element 113 on gold was claimed. The detection setup coupled to TASCA allows for studying SHE in the wide range of chemical properties, and in a direct comparison with its neighbors in the periodic table of elements. The first measurement of element 113 in a chemistry experiment at GSI was conducted in 2016. The preliminary results will be presented.

Development of Designer Molecules for Use in Future Superheavy Element Chemistry Experiments

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Our group at Texas A&M University has been studying the use of modern “designer” molecules for their potential application in superheavy element chemistry experiments. The latter has included ionic liquids (organic salts that are liquid at room temperature) [1, 2], deep eutectic solvents (DESs) [3], and low-transition-temperature materials [3]. These classes of materials allow for tremendous tunability in their properties through modification of substituents and/or variation of constituent concentrations. For example, our group has investigated the extraction of the expected element 113 homologs, In and Tl, from HCl into ionic liquids composed of substituted imidazolium bis(trifluoromethanesulfonyl)imide ionic liquids with varying alkyl chain lengths. In optimum cases, the Tl(III)/In(III) separation factor was $>10^7$ in the presence of a strong oxidizing agent, showing a substantial difference in behavior that could be exploited to better understand element 113 chemistry. Using tetraheptylammonium chloride-based DESs, distribution ratios of $>10^3$ have been observed for extraction of In from oxalic acid, and experiments with Tl are ongoing. These DESs are also more environmentally friendly than traditional solvents and are available at relatively low cost. There are also possibilities to apply these designer molecules to gas-phase experiments [2], such as using them as sorbent materials in a coated column. Finally, a new recoil transfer chamber with fast extraction has been fabricated and characterized [4]. Efficiencies of tens of percent have been measured in online experiments without the need for oscillating electric fields.

This talk will summarize these developments and the most recent results. In addition, potential new chemical experiments on element 113 will be discussed.

References

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Chemistry of flerovium - current status

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for a Mainz Univ. - GSI Darmstadt - HIM Mainz - JAEA Tokai - Liverpool Univ. - LLNL Livermore - Nevada Las Vegas Univ. - Lund Univ. - RIKEN Wako - SINP Kolkata - IITR Uttarakhand – ANU Canberra - Oslo Univ. - Jyväskylä Univ. - ITE Warsaw - collaboration
(The TASCA flerovium chemistry collaboration)

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The experimental determination of chemical properties of element 114 (flerovium, Fl) is a topic of high interest in current transactinide element research. Early atomic calculations indicated that Fl could be chemically inert [1]. More recent relativistic calculations confirm that it should be more inert than its lighter homolog Pb. Nevertheless, it is predicted to have a distinct metallic character [2].

The first experimental study to measure the properties of Fl was performed by a PSI/FLNR- collaboration without pre-separation [3]. Based on the observation of three radioactive decays attributed to Fl isotopes, a rather weak physisorption on gold $-\Delta H_{\text{ads}}^{\text{Au}}(\text{Fl}) = 34^{+54}_{-11}$ kJ/mol, was inferred [3]. The first successful chemical investigation of Fl using a pre-separator was performed at the gas filled recoil separator TASCA, at GSI, Darmstadt. Two Fl events were detected on gold at room temperature ($-\Delta H_{\text{ads}}^{\text{Au}}(\text{Fl}) > 48$ kJ/mol) with negligible background [4]. Based on this result, the formation of a metal-metal bond of Fl with gold was inferred. Limited statistics and contradicting results of these two experiments called for advanced experimental studies. Additional experiments performed by the PSI-FLNR collaboration, did not result in Fl observation [5]. Recently two advanced Fl studies were performed at TASCA. Six more radioactive decays of Fl were observed. This talk covers the status of the chemical analysis of all Fl experiments, conducted by GSI Darmstadt, using the gas-filled separator TASCA for pre-separation.

References

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Measurements with the SHIP focal plane detector system 2016

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The SHIP focal plane detector system composed of a silicon box detector for alpha and conversion electron detection, and a Ge clover detector for gamma-ray detection [1] was employed at SHIP with parasitic ⁴⁸Ca beam in June of this year. Data from neutron deficient Np isotopes were taken, complementing data acquired from the 2015 beam time at SHIP [2]. An overview of measurements will be presented, along with continuing development plans for the device.

References

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Speeding up gas-phase chemistry to access elements beyond FI

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In recent years, the chemical properties of the SuperHeavy Elements (SHE) copernicium (Cn, Z = 112) and flerovium (Fl, Z = 114) have been extensively studied. Here, the combination of the gas chromatography with the electromagnetic preseparator TASCA is an indispensable tool to obtain the required sensitivity for the single atom studies. Gas chromatographic techniques, which rely purely on flowing gas to flush the produced isotopes into the chromatography detector, are applicable to isotopes with half-lives that are longer than about 0.5 seconds. Looking at elements beyond Fl, the extraction time of typically 0.5 s with this setup is significantly longer than the half-life of the most long-lived isotopes of all elements with Z > 114. For element 115, for example, the most long-lived currently known isotope is ²⁸⁸115 with T_{1/2} = 170 (⁺⁴⁰/₋₃₀) ms. To get access to the short-lived isotopes a faster extraction technique is accordingly needed. This talk covers the combination of a gas stop cell with electric fields and the COMPACT detector. This new gas-catcher allows extraction times in the range to tens of milliseconds. A first offline experiment with this setup was performed in April 2016 and the preliminary results will be presented.

Identification of multi-nucleon transfer reaction products in ⁵⁰Ti + ²⁴⁹Cf

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At the gas-filled recoil separator TASCA, GSI Darmstadt, experiments on the synthesis of elements beyond Z = 118 have been undertaken in the past years [1]. The element with Z=120 was searched in ⁵⁰Ti + ²⁴⁹Cf → ²⁹⁹120* fusion reaction, which is predicted to yield the highest cross sections among several conceivable reactions leading to the production of this element.

At the focal plane detector of TASCA not only the evaporation residues of fusion reactions, but also transfer reaction products, which are not 100% suppressed inside any in-flight separator, can be measured [2]. Transfer products from the ⁵⁰Ti + ²⁴⁹Cf reaction were identified by different correlation analyses involving implantation signals and alpha particles that were detected with a Combined ANalog and DIgital data acquisition system CANDI [3].

The results on identified transfer products as well as the data analysis will be discussed.

References

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Pile-up pulse analysis with fast sampling ADC techniques

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Above nuclear shell closures, fast α -decaying nuclei with half-lives down to tens of nano seconds exist.

Due to their short lifetimes, it has been difficult to measure their properties as α -decays result in pile-ups with an analogue electronics experimental set-up.

For the superheavy element 115 experiment, conducted in 2012 at GSI TASCA lead by the Lund University Nuclear Structure Group, the employment of fast sampling ADCs was the case which made it possible to extract energies and times of pile-ups with tailor-made algorithms.

A pile-up trace analysis routine has been developed to extract the energies and times from the preamplifier traces and was applied to the 2012 experimental data. From the obtained results a connection to the tabulated α -decay chain of Ra-219 to Rn-215 could be firmly established.

Nuclear Spectroscopy with GEANT4: The Superheavy Challenge

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The simulation toolkit GEANT4 was originally developed at CERN for high energy physics but over the years it has been established as a *swiss army knife* not only in particle physics but it has seen an accelerated expansion towards nuclear physics and more recently to medical imaging and gamma- and ion- therapy to mention but a handful of new applications.

The validity of GEANT4 is vast and large across many particles, ions, materials and physical processes with typically different various models to choose from. Unfortunately atomic nucleus with atomic number $Z > 100$ are not properly supported. This is likely due to the rather novelty of the field, it's comparably small user base and scarce evaluated experimental data.

To circumvent this situation different workaround have been used over the years. In this work the simulation toolkit GEANT4 will be introduced with its different components and the effort to bring the software to the heavy and superheavy region will be described.

Theoretical Study of Molecular Properties of Group-VI, VII and VIII Heavy and Superheavy Metal Carbonyls

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Volatile metal carbonyl complexes of super-heavy elements are in focus of experimental investigations. To render assistance to those experiments we present results of the theoretical studies of molecular properties of group VI, VII and VIII super-heavy element carbonyls and their homologues. Properties like equilibrium geometry, polarizability, ionization potential etc. are obtained with most modern relativistic quantum chemistry methods which are at disposal in ADF, ReSpect and DIRAC program suites. Calculated values are compared with available theoretical [1] and experimental data.

References

- [1] Pershina, V. and J. Anton, J. Chem. Phys. **138**, 174301 (2013)



Studies of the thermal stability of Sg(CO)₆

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In recent years gas phase chemical studies assisted by physical pre-separation allowed for the investigation of fragile single molecular species by gas phase chromatography. The latest success revealed the formation of a highly volatile carbonyl species of the heaviest group 6 transactinide seaborgium and its adsorption interaction with quartz surfaces. We report here on preliminary results of a second generation experiment to investigate the intramolecular bond between the central metal atom of the complexes and the ligands addressing the influence of relativistic effects in the heaviest compounds. Results are shown in conjunction with a suggestion of a data analysis revealing a weaker metal-carbon bond within this complex for Sg compared to its lighter homologue tungsten. Furthermore, our study reveals the formation of a secondary, less volatile carbonyl species for Mo and to a lesser extent for W.

In-situ synthesis of carbonyl complexes with short-lived isotopes of group VI, VII and VIII elements

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The chemical investigation of the superheavy elements is of broad interest, as relativistic effects, which scale with Z^2 , are predicted to strongly influence the chemical behavior of these elements¹. Thus, trends in the periodic table might be broken. Comparative studies of the transactinides with their lighter homologs help to understand the influence of relativity on the electron shell and test the predictive power of the periodic table.

However, due to experimental limitations only a few compounds of the superheavy elements (SHE) have been studied so far. At the moment the combination of physical pre-separation with carbonyl chemistry is the method of choice. However, the fusion products from the asymmetric 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, the efficiencies are in order of 10% for Sg. Thus, the overall efficiency of the synthesis of carbonyl complexes in combination with physical pre-separation is rather low. The efficiency in the carbonyl chemistry studies becomes even more important for transactinides heavier than Sg due to a significant reduction of the production cross sections. In future experiments, the possibilities for chemical investigation of the metal carbonyl complexes of SHE without using a pre-separation stage should be explored, which would avoid the corresponding losses of 90%. First experiments performed in Tokai (JP) 2015, suggest that the successful synthesis of Os and W carbonyl complexes also without a pre-separator is feasible. The most significant result was the observation of a volatile tungsten species where no oxygen content was present. Further experiments would be performed at the TRIGA Mainz Reactor to confirm this results.